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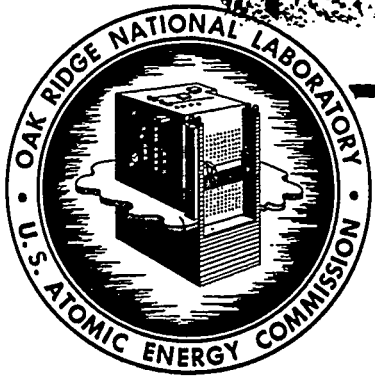
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PUREX PILOT PLANT QUARTERLY REPORT  
FOR FEBRUARY, MARCH, AND APRIL, 1951

By Authority Of:



**OAK RIDGE NATIONAL LABORATORY**  
OPERATED BY  
**CARBIDE AND CARBON CHEMICALS COMPANY**  
A DIVISION OF UNION CARBIDE AND CARBON CORPORATION

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PUREX PILOT PLANT QUARTERLY REPORT

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FOR FEBRUARY, MARCH AND APRIL, 1951

H. K. Jackson

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ORNL Pilot Plant  
February-March-April 1951 Report

#### 1.0 Abstract

This report presents the program and results of the ORNL pilot plant development of the Purex process, and presents the status of a new program for the separation of plutonium from irradiated Chalk River Fuel Rods.

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2.0 Introduction

The primary objective of the Purex Process development in the ORNL pilot plant is the demonstration of the satisfactory separation of uranium, plutonium and fission products from each other and the adequate recovery of uranium and plutonium in the pulse column contactor. Other objectives include the design and installation of equipment for the removal of radioactive rare gases from dissolver off gases; the design, installation and operation of a system for the volume reduction and simultaneous recovery of nitric acid from the hot waste IAW stream; the demonstration of a solvent recovery process; and the development of methods for processing waste streams.

A new program designated as the SCRUP Program is being initiated to recover plutonium from irradiated Chalk River fuel rods. The Purex Process will be used for this program, which will be conducted in the ORNL Metal Recovery Building now nearing completion. This program will permit testing of the Purex Process in pulse columns at a maximum throughput of 1500 pounds of uranium per day. The ORNL Metal Recovery Program has been deferred until January 1952, at which time the two programs will be run concurrently.

During the past quarter, most of the effort of the pilot plant has been directed toward stage height and flooding determinations in both packed and pulse columns using cold uranium as feed. Five hot runs were made at levels from 20 to 100 grams plutonium per ton of metal. A slug dissolution procedure was evolved for use in conjunction with the installation of rare gas recovery equipment, and the equipment for nitric acid recovery from the hot IAW stream was installed and tested. A batch process for solvent cleanup was successfully operated.

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Introduction (continued)

The future program of the pilot plant with respect to Purex Process development will consist of the installation and operation of the fumeless dissolving equipment, continued testing of the nitric acid recovery system, development and installation of a continuous solvent recovery process, and design of a waste stream treatment to include all process waste streams. Following demonstration runs at full Hanford level, the pulse columns will be operated at the 100 g/t level to obtain data on decontamination and losses as functions of the pulse column variables such as pulse amplitude, pulse frequency, volume velocity, plate hole diameter, plate spacing, percentage free area and pulse shape.

3.0 Summary

A program of cold runs using packed columns was completed during the past quarter, and a program of pulse column runs using both inactive and active feeds was started.

During the packed column program it was found that the stage height using 1/4 inch x 3/8 inch Raschig ring packing was relatively high; the average HETS values for uranium in the IA, IB and IC columns were 6.2 feet, 5.2 feet and 9.8 feet. The flooding rate was found to be low, approximately 100 gal/sq. ft./hr (both phases) in the IA column.

In pulsed columns the stage height for uranium was decreased by a factor of about two for the IA and IB columns and by a factor of about four for the IC column; the average HETS values obtained were 2.8 feet, 3.1 feet and 2.6 feet for the IA, IB and IC columns. A pulse amplitude of 0.9 inch and a pulse

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Summary (continued)

frequency of 58 strokes per minute were used for most of the IA column runs, and under these conditions flooding in the IA pulsed column occurred at 600 gal/hr/sq. ft. For both pulsed and packed columns flooding occurred in the scrub section.

The initial hot runs in pulse columns were made using a 0.5 inch amplitude in the IA column scrub section, and at this amplitude the stage height for fission products was about four feet. The stage height was reduced to two feet by increasing the pulse amplitude to 0.9 inch. Under these conditions, first cycle beta decontamination factors of  $2 \times 10^4$  were obtained for uranium and plutonium.

Uranium through two pulse column cycles was decontaminated to seven parts per billion of plutonium and to a fission product beta activity one percent of the background of natural uranium. A plutonium fission product beta decontamination factor of  $10^6$  through two cycles was demonstrated. Steady state conditions yielded 0.15 parts of uranium per part of plutonium.

The installation of acid recovery equipment was completed, and the results of cold runs indicated that product acid concentrations of 65 percent could be obtained with column losses of 1.5 percent. The IAW stream from one 50 g/t run was processed. This stream was volume reduced by a factor of twenty, and decontamination factors for the recovered acid and the condensate were  $4 \times 10^5$  and  $2 \times 10^6$ . No volatilization of ruthenium could be detected.

Preliminary design was begun for the recovery of radioactive rare gases liberated during dissolving. The process now under consideration consists of the removal of iodine, oxides of nitrogen and carbon, hydrogen and moisture, and the subsequent adsorption of radioactive xenon and krypton. Instrumentation

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Summary (continued)

for automatic temperature control of the dissolver has been installed to obtain a more constant flow of off-gas.

4.0 Purex Process Development

Brown, Barclay, Connair, Darby, Farrow, Harrington, Hill, Hylton, Hull, Landry, Lewis, Lindauer, Parker, Porter, Sadowski, Spencer, Ullmann, Watkins, Winget, Wood, Yancey

4.1 Dissolving and Feed Preparation

4.11 Automatic Control of Dissolver

During the Purex conversion the dissolver and its instrumentation were modified to permit automatic control during the dissolving of uranium metal. This was done to reduce the operator labor required for dissolving and also to obtain a more constant flow of gas to that part of the system where the radioactive rare gases will be separated. Instrumentation was provided to control the vacuum in the dissolver and to control the temperature of dissolving. The dissolver vacuum is measured, recorded, and controlled by a Taylor aneroid controller, which actuates a 1-1/2 inch diaphragm operated air valve in the off-gas line. The dissolver is maintained at six inches of water vacuum. The temperature of the dissolver solution is controlled by a Brown Electronic Potentiometer with air control. The output air from this instrument is applied to two Mason-Neilan split range valve positioners, which control the flow of steam and water to the dissolver jacket. The temperature of the dissolver is maintained to  $\pm 2^{\circ}\text{C}$ . With a dissolver



Automatic Control of Dissolver (continued)

equipped with coils instead of a jacket the temperature of dissolving could probably be controlled to  $\pm 0.5^{\circ}\text{C}$ . In the pilot plant dissolving the temperature is set at  $90^{\circ}\text{C}$  for the first two hours,  $95^{\circ}\text{C}$  for the next two hours, and at  $105^{\circ}\text{C}$  until the desired specific gravity is reached.

4.12 Plans for Fumeless Dissolving

The radioactive rare gases liberated during dissolving at the Savannah River plant must be separated and held in storage indefinitely. The process now under consideration consists of the removal of iodine, nitric oxide, nitrogen dioxide, nitrous oxide, carbon monoxide, carbon dioxide, hydrogen, moisture and the subsequent adsorption of radioactive xenon and krypton.

A dissolver off-gas treatment system which consists of the following principal units is being designed for the Pilot Plant: (See Fig. 2).

1. An electrical gas heater followed by an adsorption tower packed with silver nitrate coated Berl saddles for the removal of iodine.
2. A water seal chamber to prevent the backup of oxygen into the dissolver.
3. A primary gas holder of the liquid displacement type and approximately one fourth the total volume of the gases evolved. Oxygen will be metered into the holder to oxidize nitric oxide to nitrogen dioxide which will then be absorbed by the displacing fluid. Only about 95 per cent of the total oxygen required for complete oxidation will be added at this point.

Plans for Fumeless Dissolving (continued)

4. A secondary gas holder of the liquid displacement type into which the residual gases from the primary holder will be transferred. The remainder of the oxidation of nitric oxide and absorption of nitrogen dioxide will be accomplished in this secondary holder. Oxidation can be either by manual addition of oxygen or use of 3 per cent hydrogen peroxide.
  5. Residual gas in the secondary unit will be displaced through a nitrous oxide removal unit. This consists of either catalytic decomposition of the nitrous oxide, reacting the nitrous oxide at 600-800°C with copper to yield cupric oxide and nitrogen, or combustion of the nitrous oxide with either hydrogen or carbon monoxide.
  6. The gases will then be passed through a tower containing either caustic pellets or pebble lime to remove any residual carbon dioxide or nitrogen dioxide.
  7. A hot copper oxide tower will be required to remove excess carbon monoxide or hydrogen if these gases are used for combustion with nitrous oxide (see item 5). This unit may be required in any case if either of the above gases are present to any extent from the dissolution reaction.
  8. A condenser followed by an alumina dryer will be used to reduce the moisture content of the gas stream.
  9. Xenon and krypton will be adsorbed at approximately the temperature of liquid nitrogen onto either silica gel or activated alumina. The residual gas, mostly nitrogen, will be exhausted from this unit to the stack.
- [REDACTED]
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#### 4.13 Operational Variables and Recommended Procedures

The purpose of the initial Purex dissolvings was the production of a suitable feed for the solvent extraction process. Increased interest in fumeless dissolving made it necessary to modify both equipment and dissolving procedure in order to prepare the pilot plant for the installation of fumeless dissolving equipment. The non-irradiated metal dissolvings (Table 1) represent the partial evolution of a dissolving procedure which is adaptable to fumeless dissolving. The ORNL irradiated slug dissolvings (Table 2) resulted in a rapid, safe dissolving method which should require almost no change when the fumeless dissolving equipment is installed.

The principal steps in the process beginning with the slugs and ending with final first cycle feed adjustment are these: (1) coating removal, (2) nitric acid dissolution of uranium, (3) feed adjustment, (4) filtration and (5) final feed adjustment.

##### 4.131 Coating Removal

The aluminum jacket on an ORNL slug weighs 24.5 grams. This jacket must be entirely removed prior to dissolution of the uranium metal in nitric acid. The pilot plant is following the established procedure for jacket removal which consists of digesting the slugs in a 2.7M sodium nitrate - 2.8M sodium hydroxide solution for one hour at 104°C. The sodium hydroxide to aluminum molar ratio should be above 1.6 to prevent precipitation of the sodium aluminate formed. The volume of solution used in the pilot plant is the minimum volume required to cover the slugs; this volume gives a sodium hydroxide/aluminum

Coating Removal (continued)

ratio of three.

A modified coating removal procedure is required for Hanford slugs. The aluminum jacket on a four-inch Hanford slug weighs approximately 65 grams, and much of this aluminum is found on the ends of the slugs. These slug end caps are not completely dissolved or even detached from the slugs by the ORNL slug coating removal procedure. The pilot plant will use the following procedure for Hanford slug coating removal: the solution will be 2.7M with respect to sodium nitrate, 4.0M with respect to sodium hydroxide, and the volume of solution will be such that the caustic to aluminum molar ratio will be three. The digestion time at 104°C will be four hours. Two consecutive coating removal steps will be made on Hanford slugs until a procedure has been developed which will remove all the aluminum in one operation.

4.132 Nitric Acid Dissolution of Uranium

Among the variables encountered in uranium slug dissolving are the following: (1) amount of metal heel remaining in the dissolver, (2) quantity of air pulled through the dissolver, (3) initial molar ratio of nitric acid to uranium which is to be dissolved, (4) initial concentration of nitric acid in the dissolver, (5) water addition through the condenser ring spray, (6) temperature regulation during dissolving, (7) the dissolver pressure, and (8) the specific gravity to which the dissolving is allowed to progress.

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S. T. C. [REDACTED]

Amount of Metal Heel Remaining in the Dissolver

The amount of heel which should remain in the dissolver after a dissolving is a function of dissolver freeboard, cooling capacity of the dissolver, permissible pressure buildup during the dissolving, the time cycle necessary to prepare feed to supply the solvent extraction operation, and chemical considerations involving valence state, colloid formation and solvent extractability.

The pilot plant dissolves to a 200 percent metal heel initially and then to a 100 per cent heel with a dissolver freeboard of 100 per cent. This is the minimum freeboard necessary because of frothing during the height of reaction, and it is believed that a freeboard of at least 200 percent would be desirable for this type of dissolving.

The time cycle is shortened by dissolving to a 200 percent heel, then to a 100 percent heel; this saves one coating removal step over dissolving to a 100 per cent heel. For ORNL slugs this is a saving of about 20 percent in the thirty-six hour overall cycle. For Hanford slugs the time saving amounts to about 30 percent for the cycle.

Quantity of Air Pulled Through the Dissolver

The first four cold dissolvings were made with no special attempt to make the dissolver air tight (Table 1). Dissolvings ISP-3a and ISP-3b were quite smooth with no tendency toward development of pressure in the dissolver. The acid utilization was only forty percent which indicated that the nitrogen dioxide formed by the air oxidation of nitric oxide was not absorbed in the

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Quantity of Air Pulled Through the Dissolver

updraft condenser. Dissolvings ISP-10a and ISP-10b were made with the presence of air in the dissolver and with a downdraft condenser through the top of which water was added. The acid utilization rose to 72 per cent indicating considerable nitric oxide oxidation and nitrogen dioxide absorption in the water.

Beginning with the first hot dissolving (Table 2), the dissolver and accessories were made as nearly air tight as possible. Due to the absence of air to oxidize nitric oxide to nitrogen dioxide which is absorbable in water, the acid utilization dropped to 40 percent. It was noted that the dissolver developed pressure quite rapidly on the 200 percent heel dissolving with high initial vacuum on the dissolver and continuing high off-gas vacuum. This was attributed to froth formation which was carried to the packed entrainment separator.

Initial Molar Ratio of Nitric Acid to Uranium

The mols of nitric acid to be charged per mol of uranium to be dissolved is dependent upon the amount of air admitted to the dissolver, the final desired acid concentration of the solution in the dissolver, and whether or not water is added through the ring spray to absorb the nitrogen dioxide formed during the reaction. Since no air is admitted during the hot dissolvings, the acid utilization is not particularly good and therefore a high initial mol ratio of nitric acid to uranium is necessary to give the desired nitric acid concentration in the final dissolver solution. At 5.0 mols initial nitric acid to 1.0 mol uranium in the dissolver, the residual acid concentration at the desired specific gravity is about 0.4M which is sufficient acid concentration to have insured a rapid

Initial Molar Ratio of Nitric Acid to Uranium (continued)

reaction rate throughout the dissolving and to prevent the formation of Pu (IV) polymer which is not extractable.

Initial Concentration of Nitric Acid in the Dissolver



The primary consideration which governs the initial concentration of nitric acid to be charged to the dissolver is the rate of dissolution of the uranium in nitric acid. Using 55 percent nitric acid dissolvings are completed in approximately eight hours.

Water Addition through the Condenser Ring Spray

There is considerable nitrogen dioxide formed in the reaction of 55 percent nitric acid with uranium even in the absence of air. Some increase in acid utilization can be achieved by the absorption of nitrogen dioxide in the water from the top of the condenser. This effect will be measured during future dissolvings.

Temperature Regulation During Dissolving

The rate of reaction of uranium slugs and nitric acid is quite slow under 70°C, but between 70°C and 80°C there is a marked increase in the rate of reaction. In the early dissolvings the temperature of dissolving was controlled to 102°C. As long as air was admitted to the dissolver, the dissolver did not go from vacuum to pressure. With the exclusion of air and the 102°C temperature control point, the dissolver went from vacuum to pressure.

Temperature Regulation During Dissolving (continued)

By controlling the temperature to 90°C during the first two hours of dissolving, to 95°C for the next two hours, and then setting the control to 105°C until the desired specific gravity is reached, a uniform vacuum dissolving can be made.

Dissolver Pressure

Most of the dissolvings have been made with the dissolver under six inches of water vacuum. The final fumeless dissolving design may make it necessary to lessen the vacuum on the dissolver or even to carry the dissolver under a slight pressure.

The Specific Gravity to Which the Dissolving Progresses

The final specific gravity to which the dissolving progresses is determined primarily by the uranium and nitric acid concentration required in the final feed and such practical considerations as keeping the solutions from crystallizing while cooling to jet, the number and amount of the necessary jet dilutions before final feed adjustment, and the time required to gain a given specific gravity increment in the dissolver. For the pilot plant system, a final specific gravity of 1.73 at 105°C was found to be satisfactory. At this specific gravity the uranium concentration is 560 g/l, and it is necessary to dilute the dissolver solution to 460 g/l of uranium prior to cooling to 40°C (jetting temperature) to prevent precipitation of the uranyl nitrate.



#### 4.133 First Cycle Feed Adjustment



The preparation of feed for one solvent extraction run requires two dissolvings which are combined in the initial feed preparation tank along with a dissolver rinse. There are two operations to be carried out in the primary adjustment tank: (1) The uranium and nitric acid content of the solution must be so adjusted that the treatment in the final adjustment tank will consist of the addition of a small amount of water and nitric acid. (2) The plutonium must be adjusted to the tetravalent state by sodium nitrite addition.

##### Uranium and Nitric Acid Adjustment

An evaporation of the dissolver solutions is sometimes necessary in the feed adjustment tank due to the dissolver rinse and a variable jet dilution. The time order of the evaporation and nitric acid addition is determined by plutonium behaviour. In low nitric acid concentration and particularly in thermally hot solutions, plutonium (IV) tends to disproportionate to the trivalent and hexavalent states. This indicates that the nitric acid addition should be made before evaporation. This disproportionation does not occur in the dissolver due to the presence of the metal heel.

##### Plutonium Valence State Adjustment

Most of the plutonium in the feed adjustment tank is present as the tetravalent plutonium although some trivalent and some hexavalent plutonium is present. Sodium nitrite is added to give a concentration of 0.02M in the



Plutonium Valence State Adjustment (continued)

final feed solution. The feed solution is digested for two hours at 70°C to bring about two reactions. Trivalent plutonium is oxidized to the tetravalent state and hexavalent plutonium is reduced to the tetravalent state by sodium nitrite treatment.

4.134 Filtration

It has not been definitely demonstrated that a filtration or centrifugation of feed solutions from either ORNL or Hanford slugs is necessary when the pulse column is used for a contactor. At present the pilot plant is filtering the feed prepared from both types of slugs through a sintered stainless steel 65 micron filter. An aqueous wash of the filter following feed filtration prevents significant uranium and plutonium losses during filtration.

4.135 Final Feed Adjustment

If the preceding steps in feed preparation have been properly carried out, the final feed adjustment consists only of the addition of a small volume of water and nitric acid. The final uranium concentration is adjusted to  $320 \pm 10$  grams per liter and the nitric acid adjusted to  $2.0 \pm 0.1M$ .

4.2 Solvent Extraction

4.21 Columns and Accessories

4.211 First Cycle and Second Uranium Cycle Packed Columns

Packed columns and accessories remain essentially unchanged from three months ago. Column dimensions and heights are given in

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First Cycle and Second Uranium Cycle Packed Columns (continued)

the following table.

Packed Column Dimensions

<u>Column Section</u>	<u>Diameter</u>	<u>Packed Height</u>
IA Extraction	3"	19'
IA Scrub	3"	21'
IB Partitioning	3"	28'
IB Scrub	3"	13'
IBs Scrub	2"	15'
IC Strip	4"	36.5'
ID Extraction	3"	20'
ID Scrub	3"	20'
IE Strip	4"	39'
Columns packed with 1/4" x 3/8" Raschig Rings		

Since all columns have been lengthened to the maximum height of the cells, cascade flow between columns is not possible. Instead the IAP, IBU and IDU streams are lifted by air jets to elevated accumulator pots whence they flow by gravity to the succeeding columns.

The only packed column changes from the originally designed columns were:

- (1) Increasing the diameter of the A and D columns from 2 inches to 3 inches to increase the capacity to 50 kg uranium per day.
- (2) Increasing the height of the C and E columns from 21 feet to 36.5 feet and 39 feet respectively to reduce stripping losses.

4.212 First Cycle and Second Uranium Cycle Pulse Columns

During the past three months pulse column runs have been made under conditions given in the following table.

Pulse Column Dimensions and Conditions

<u>Column Section</u>	<u>Diameter</u>	<u>Packed Height</u>	<u>Amplitude</u>	<u>Frequency</u>
IA Extraction	2"	7.5', 8.5'	0.62" to 1.08"	58
IA Scrub	2", 3"	8.0', 12.5'	0.27" to 0.92"	58
1B Partitioning	2-1/2"	8.0', 11.5'	0.43" to 0.86"	58
1B Scrub	2"	6', 8'	0.62" to 1.23"	58
1C Strip	4"	12'	0.42" to 0.56"	73
1D Extraction	2"	8', 8.5'	1.08"	58
1D Scrub	2", 3"	8', 8.5'	0.48"	58
1E Strip	4"	15'	0.49" to 0.56"	73

The perforated plates for each column have 1/8 inch-diameter holes, 23 percent free area and are spaced 2 inches apart. Gravity flow between columns is used and each column is pulsed with a Milton Roy pump located in a cold area 15 to 25 feet from the columns. At the pulse frequencies now being used, the capacity of the system is limited by the diameter of the IA column scrub section. Using a 3 inch scrub section the maximum throughput is 175 kg of uranium per day.

Subsequent to the completion of runs covered in this report the columns were modified to provide greater flexibility. The 1A and 1B columns were lengthened to 30 and 26 feet respectively and 1C column was modified to permit operating with either aqueous or organic phase continuous. The following

First Cycle and Second Uranium Cycle Pulse Columns (continued)

combinations of scrub, extraction and partitioning sections are now possible in the IA and IB columns.

Column	IA		IB	
	Scrub	Extraction	Partitioning	Scrub
Height of section, feet	20	10	18	8
	18	12	16	10
	12	18	14	12

4.213 Second Plutonium Cycle Equipment

The equipment is located in Building 3503, and was formerly used for the 23 Process development. A schematic equipment flowsheet is given in Figure 5. The process tanks range in capacities from 20 to 100 gals. and are equipped with accessories for mixing, heating and cooling, liquid level and specific gravity determinations, overflow alarms, and temperature measurement. Process solutions are transferred by pumps, steam jets, vacuum and gravity and are sampled by recirculating air jet samplers. Makeup tanks outside the cell have capacities up to 50 gallons.

Feed Preparation

The IBsP stream after being volume reduced by a factor of five is transferred in casks to the second plutonium cycle equipment. The solution is drawn by vacuum to a feed preparation tank, and adjusted to flowsheet specifications for nitric acid and sodium nitrite. At the end of the valence adjustment step the feed is transferred by vacuum and gravity into the feed tank. From this point

Feed Preparation (continued)

it is pumped to the column by displacement.

Solvent Extraction

The IIA extraction column is a 1-1/2 inch stainless steel pipe packed with 1/4 inch by 3/8 inch stainless steel Raschig rings. Alternate feed points in a packed height of 41.5 feet permit wide variation of the extraction to scrub height ratio.

The IIB stripping column is a 2 inch stainless steel pipe packed with 1/4 inch by 3/8 inch Raschig rings. Three feed points are provided in a packed height of 40 feet.

The IIBs scrub column is a 27 feet, 1-1/2 inch stainless steel pipe packed with 1/4 inch by 3/8 inch Raschig rings. The purpose of this column is to remove the last traces of TBP in the Pu stream by contacting with fresh diluent.

The interface on each column is maintained by a Taylor instrument controlling the air pressure to a jackleg pressure pot.

4.22, Summary of Cold Column Runs

Thirty-six runs using cold uranium feed were made in packed and pulsed columns to test the Purex Flowsheets No. 1 and No. 2 (see Figs. 12 and 13). As the program progressed several changes in column dimensions were made to increase capacity and reduce losses. Since most of the features of the extraction, partitioning and strip columns are similar in both cycles and both flowsheets, the summary data presented below is organized on the basis of columns by function regardless of flowsheet or cycle. Detailed data for these runs are presented

Summary of Cold Column Runs (continued)

in Tables 3 through 9.


4.221 Results of Packed Extraction Column Cold Runs

This paragraph summarizes the results of IA and ID column runs using Flowsheet No. 1, and IA and IIA column runs using Flowsheet No. 2.

Flooding in the A column started at 100 gal/hr/sq. ft. (both phases). Some runs were completed at flowrates up to 160 gal/hr/sq. ft., but column operability at this rate was not satisfactory. The phenomenon observed was that the AW flow was doubled for a period and would then cease while the interface dropped until sufficient aqueous was introduced into the column to raise the interface. The organic apparently collected in the column displacing aqueous from the bottom of the column at an abnormally high rate. The organic then rose in a slug to yield an abnormally low interface. After an hour the interface was restored to the top. The sequence of events was cyclical with a frequency of the order of two hours. The average HETS and HTU values at 100 gal/hr/sq. ft. in the scrub section were 6 ft. and 2 ft. This corresponds to an average uranium loss of 0.01 percent in 19 ft. of extraction section.

4.222 Results of Packed Partitioning Column Cold Runs (IB,IIB)

"B" column runs were made at flowrates from 45 to 150 gal/hr/sq. ft. in the partitioning section without evidence of flooding. The average HETS and HTU values for uranium were 5 feet and 1.6 feet. Uranium losses of 0.01 percent were obtained in 13 ft. of scrub section. Losses did not change significantly when flowrates were varied by a factor of three.



4.223 Results of Packed Strip Column Cold Runs (IC, IIC, IE)

The "C" column was operable at rates up to 400 gals/hr/sq. ft. (both phases). The ICS diluent solvent stream (introduced into the IAP to lower its density) was found unnecessary. A 0.02 M nitric acid strip showed no advantage over a water strip. A three fold reduction in losses was obtained by an increase of the aqueous to organic strip ratio from 3:2 to 2:1. Further increase in strip ratio yielded no improvement in losses. Losses increased by a factor of five when the flowrate was decreased from 400 to 200 gal/hr/sq. ft. For the 3:2 strip ratio average HETS and HTU values were 10 feet and 7 feet corresponding to 2 percent losses in 37 feet of column. The two-column cycles produced higher stripping losses than the three-column cycles because of the dependency of the equilibrium upon the nitric acid concentration in the entering organic stream.

4.224 Results of Pulsed Extraction Column Cold Runs

Flowrates up to 1095 gals/hr/sq. ft. in the extraction section of the "A" column were used without any indication of flooding. In the scrub section flooding occurred at 600 gals/hr/sq.ft. with a 0.5 inch amplitude and 58 strokes per minute. HETS and HTU average values were 3 feet and 0.9 feet corresponding to a uranium loss of 0.01 percent in 8.5 feet. The losses to the AW were steady over a three fold range of throughput and a 20 percent variation in pulse amplitude.

4.225 Results of Pulsed Partitioning Column Cold Runs (IB, IIB)

No flooding was observed at rates up to 780 gals/hr/sq. ft.



Results of Pulsed Partitioning Column Cold Runs (IB, IIB) (continued)

through the partitioning section with a 0.9 inch amplitude and 58 strokes per minute. Uranium losses in the scrub section were steady over a three fold range of throughput and a two-fold range of pulse amplitudes. Average HETS and HTU values were 3 feet and 1 foot corresponding to 0.02 percent uranium loss in 6 feet of scrub section.

4.226 Results of Pulsed Strip Column Cold Runs (IC, IIC)

No flooding was observed at rates up to 615 gals/hr/sq. ft. as long as the amplitude was held below 0.5 inch. Increased amplitude lowered losses but tended to eject some organic into the ICU stream. Average HETS and HTU values were 2.6 feet and 1.7 feet corresponding to 0.5 percent uranium loss in 12 feet of column. Increase of the aqueous to organic strip ratio from 3:2 to 5:2 reduced losses by a factor of three.

4.227 Miscellaneous Observations

- (a) IBs column operation was unsatisfactory when 1/4 inch x 1/4 inch Raschig rings were employed. 1/2 inch x 1/2 inch packing was installed.
- (b) Bleeding of pulse pumps to remove entrapped air improved column operation.
- (c) In the No. 2 Flowsheet runs appreciable corrosion of the Type 347 stainless steel evaporator was found. Two-thirds of the fluoride in the IIBF distilled into the condensate during the five fold volume reduction.

Miscellaneous Observations (continued)

- (d) A tan coloration was observed in solvent which had been in contact with nitrite solutions. No effect on uranium losses was noted.
- (e) The degree of aqueous entrainment from pulse columns was 2.0 percent in the IAP and 0.7 percent in the IBU.

4.23 Summary of Hot Column Runs

Feeds of increasing activity level have been processed through two cycles of uranium and plutonium decontamination and recovery.

4.231 Flowsheet #1 Hot First Cycle Pulse Column Runs

Five runs have been conducted in which feed was made from slugs irradiated in the ORNL pile and cooled a minimum of seventy days.

A summary of the data is presented in Table 10. Points noted are:

- (a) With a flowrate of 460 gals/hr/sq. ft. and an amplitude of 0.92 inch gross beta decontamination in the IA scrub section was improved compared to a flowrate of 340 gals/hr/sq. ft. and an amplitude of 0.41 inch. This change did not alter the ruthenium beta decontamination significantly but increased the decontamination of zirconium beta by a factor of 40, columbium beta by a factor of 5, total rare earth beta by a factor of 30, and gross gamma by a factor of 2. The net effect by analogy to batch countercurrent studies was an increase in the number of scrub stage from three to six without any altered column height. A gross beta decontamination factor of  $3 \times 10^3$  over the

Flowsheet #1 Hot First Cycle Pulse Column Runs (continued)

IA column was obtained in 12.5 feet of scrub section.

- (b) The bulk of the zirconium carried into the IAP entered the aqueous phase in the IB column while the bulk of the ruthenium remained in the organic. At a throughput of 390 gals/hr/sq. ft. in the IB partitioning section and an amplitude of 0.75 inch, a gross beta decontamination factor of  $1 \times 10^4$  and a gross gamma decontamination factor of  $4 \times 10^3$  were obtained for the IBP.
- (c) In the IC column with the aqueous phase continuous a ruthenium decontamination factor of six was obtained consistently and yielded for the uranium through the cycle decontamination factors of  $2 \times 10^4$  and  $4 \times 10^3$  for gross beta and gross gamma respectively.
- (d) In the runs in which only three scrub stages were attained in the IA scrub section, 40 percent of the  $UX_1 + UX_2$  in the feed entered the IBP. In the run which yielded six scrub stages, however, 90 percent of the  $UX_1 + UX_2$  entered the IAW.
- (e) Uranium losses were 0.06 percent and Pu losses (by TFA method) 0.6 percent in 7.5 feet of IA extraction section.
- (f) Uranium losses in 6 feet of IB scrub section were reduced to below 0.001 percent at a rate of 190 gals/hr/sq. ft. and an amplitude of 1.08 inches.
- (g) Plutonium losses in 11.5 feet of IB scrub section were reduced to 0.4 percent at a throughput of 390 gals/hr/sq. ft. and an amplitude of 0.75 inch.

Flowsheet #1 Hot First Cycle Pulse Column Runs (continued)

- (h) Uranium losses of 0.5 percent were obtained in 12 feet of IC column operated with the aqueous phase continuous.
- (i) Reduction of the IBsX stream by a factor of two caused no apparent difference in column IBs operation.
- (j) Batch studies indicated that the plutonium losses in the IA and IB columns could be reduced by additional contacting. Plutonium losses to the IBU were reduced by contact with 0.6M nitric acid to the same extent as by IBX indicating that mass transfer and not chemical reaction kinetics was limiting.
- (k) Several IAP and IBU samples were centrifuged to determine the extent of entrained beta and alpha activity. The results were altered by less than 25 percent.
- (l) At the conclusion of the fifth run the Amsco displacement fluid was sampled. It contained  $60 \alpha$  cts/min/ml and  $4 \times 10^3 \beta$  cts/min/ml.
- (m) Average HTU values for uranium were 1.0, 1.1 and 1.4 feet in the IA extraction section, IB scrub section, and IC column respectively.

4.232 Flowsheet #1 Hot Second Uranium Cycle Pulse Column Runs

Four runs were made utilizing the adjusted ICU from the first cycle hot runs (Table 11). It was observed that:

- (a) A uranium product was produced in two cycles decontaminated from  $\beta$  activity by a factor of  $3 \times 10^5$  and from  $\gamma$  activity by a factor of  $8 \times 10^4$ . The product contained 10 parts per billion of Pu and possessed a fission

Flowsheet #1 Hot Second Uranium Cycle Pulse Column Runs (continued)

product beta activity 2 percent that of natural uranium.

- (b) IDW uranium losses of 0.003 percent were attained in 8.5 feet of ID extraction section at a rate of 730 gals/hr/sq. ft. and an amplitude of 1.00 inches. The average uranium HTU for the ID extraction section was 0.79 feet.
- (c) With flowsheet conditions IEW uranium losses were 1 percent in 15 feet of IE column.
- (d) The addition of ferrous sulfamate to the IDS stream apparently increased plutonium decontamination by a factor of about six.
- (e) A decontamination factor of five was obtained for ruthenium in the IE column.
- (f) Decontamination factors for Zr and TRE were in the range of 1 to 5 while a Cb decontamination factor of 50 was obtained through the cycle.
- (g) Reduction of the solvent TBP concentration to 27 percent increased IDW losses to 6 percent.

4.233 Hot Second Plutonium Cycle Packed Column Runs

Nine runs were made from five first cycle run IBSP products of varied activity level and degree of decontamination. The tabulated results are presented in Table 12. It was observed that:

- (a) Decontamination factors increased with increased activity of the feed. Decontamination factors for gross beta and gross gamma of 120 and 160 yielded plutonium decontamination factors through two cycle of  $1.3 \times 10^6$
- [REDACTED]

Hot Second Plutonium Cycle Packed Column Runs (continued)

- (b) IIAW plutonium losses below 0.02 percent were obtained with 16 feet of extraction section using a 2:6:12 AS:AF:AX ratio and with 30 feet of extraction using a 2:6:10 ratio.
- (c) Reduction in IIBW plutonium losses from 0.55 percent to 0.14 percent resulted from the use of 0.1M  $\text{HNO}_3$  strip instead of water.
- (d) Column IIBs plutonium losses were below 0.02 percent.
- (e) Increase of the IIA scrub section from 14 feet to 24 feet in Run IIHP-3 improved decontamination by a factor of 1.5.
- (f) The average overall  $\text{UX}_1 + \text{UX}_2$  decontamination factor obtained was 3.
- (g) For most of the runs, Zr was the chief fission product in both the feed and final product.
- (h) Decreasing the organic to aqueous ratio in IIA column yielded improved decontamination.
- (i) A run was made employing IBSP which was not volume reduced. There were no large changes in gross beta decontamination or stripping losses.
- (j) Flooding rates for the IIA and IIB columns packed with 1/4 inch by 3/8 inch Raschig rings were 860 and 560 gals/hr/ft.<sup>2</sup> (both phases) respectively. No column difficulties have been observed with operation at 40 percent of flooding.
- (k) Since six to twelve hours were required for equilibrium to be reached, in general the latter half of each run yielded superior losses and decontamination.

#### 4.24 Column HTU and HETS Results

HTU and HETS values for uranium transfer were calculated to correlate the loss data of runs made in packed and pulsed columns. The averages are as follows:

	Packed Column		Pulse Column	
	HETS (ft.)	HTU (ft.)	HETS (ft.)	HTU (ft.)
(a) IA Extraction	6.2	1.9	2.8	0.87
(b) IB Scrub	5.2	1.6	3.1	0.98
(b) IC Strip	9.8	6.8	2.6	1.7
(a) Based on aqueous phase		(b) Based on organic phase		

Since in a pulse column conditions more closely approach continuous transfer than transfer of the order of a theoretical stage per plate, HTU is a more appropriate means of representing the data than HETS.

Equations were derived (page 77) for the Number of Overall Aqueous Transfer Units for the IA extraction and IB scrub sections. They are:

$$\text{IA Extraction: } (NTU)_{OA} = 3.22 + 1.03 \ln \frac{9.24 + 0.0294 y_1}{y_1}$$

where  $y_1$  = IAW U concentration (g/l)

$$\text{IB Scrub: } (NTU)_{OA} = 1.05 \ln \frac{28.3 + 0.05 y_1}{y_1}$$

where  $y_1$  = IBP U concentration (g/l)

A plot of ICW uranium concentration vs.  $(NTU)_{OO}$  for the IC column is presented in Figure 10.

Equilibrium data reported in KAPL-461 were utilized in the computations. Operating line equations were derived from flowsheet compositions and volumes. The results show that the height required in a pulse column is lower than that of a packed column by factors of two to four for comparable losses. Since the

Column HTU and HETS Results (continued)

flooding point is higher in pulse columns, their advantage over packed columns is increased.

The HTU values from the 2 inch IA pulse column compare well with results obtained by Hanford with a 3 inch column and are about 20 percent lower than preliminary Hanford 5 inch column results. The values for the 2 inch IB pulse column are about 30 percent lower than Hanford estimated HTU's for a 5 inch column. The results from the 4 inch IC pulse column agree with Hanford results from 3 inch and 8 inch columns operated with the aqueous phase continuous.

A plot was made (Figure 11) of the effect of CX/BU strip ratio upon the number of stages required in the IC column. The rapid initial descent of the curve explains the marked effect of strip ratio upon losses observed in several runs.

A significant decrease in HTU for the IC column was observed in varying the pulse amplitude from 0.42 to 0.56 inch but otherwise no large variation of HTU with amplitude was observed. The data from hot runs will be examined in order to yield plutonium and possibly fission product HTU values.

4.25 Column Startup and Shutdown

During the cold runs a number of special samples were drawn in order to determine the transient behavior of uranium and  $\text{HNO}_3$  in packed and pulse columns. From these data, startup and shutdown procedures for hot runs were evolved and tested.



Column Startup and Shutdown (continued)

The main observations made were:

- (a) At the conclusion of a packed column run all pumps were shut off for four hours. The system was started again and it was found that concentrations had not deviated significantly from steady state values.
- (b) Seven to eight aqueous column IA changes were required to insure steady state conditions throughout the system with respect to uranium and nitric acid beginning with packed or pulse columns free of uranium. Higher IAW uranium losses were sustained during startup than at equilibrium.
- (c) Acid concentrations with and without feed were as follows:

Stream Acidity, N	IAP	IAW	IBU	IBP	ICU	ICW
With Feed	0.22	2.2	0.03	1.25	0.02	0.00
Without Feed	0.65	0.33	0.25	1.70	0.15	0.01

- (d) Shutdowns were performed in a IA, IC system with only IAX and ICX pumps running. The IAP decreased rapidly in uranium, rose to 0.75M nitric acid and then fell off slowly in acidity. The IAW uranium concentration increased by a factor of approximately twenty and then fell off again while the acidity decreased to 0.5M. The ICU acid concentration reached 0.35M as its uranium concentration fell and the ICW uranium concentration was steady.
- (e) A shutdown of IIA, IIB, IIC system (second cycle in the ORNL #2 Flowsheet) was conducted with only the IIAX, IIBS and IICX pumps running. Uranium concentrations decreased in the IIAP and IIBU streams

Column Startup and Shutdown (continued)

and increased in the IIAW, IIBP, IICU and IICW streams. Nitric acid concentrations increased in the IIAP, IIBU, IIBP and IICU streams.

From the above facts and a knowledge of the strong dependence of decontamination upon the degree of solvent uranium saturation a hot run procedure was developed to minimize plutonium losses and fission product contamination of the product streams. It embodies (1) starting the columns in the absence of feed to produce a high acid B column, (2) turning on cold UNH feed to establish steady state U and  $\text{HNO}_3$  concentrations (3) switching to hot feed to make the run proper (4) switching to cold UNH feed to clean the column of plutonium and fission products and (5) cleaning the columns of uranium by running all pumps but the feed.

In the hot runs it was found that the procedure was successful in every respect except IAW losses which sometimes increased during shutdown. Calculation of losses is based upon the period from start of hot feed till the end of cold shutdown feed. Decontamination of the uranium product is corrected for the addition of cold feed.

4.3 IAW Evaporation and Acid Recovery

The function of the acid recovery unit is to reduce the volume of the aqueous raffinate from the IA column and to recover  $\text{HNO}_3$  for reuse in the dissolver. The results of two cold runs and two hot runs indicate that the acid recovery scheme is entirely feasible. Recovered acid concentrations of 65% can be produced, with decontamination factors greater than  $10^5$  being obtained.

4.31 Equipment and Instrumentation

4.311 Equipment

A schematic flowsheet of acid recovery equipment is given in Figure 6. The principal parts are:

- (a) The evaporator (E-1) is a conical-bottomed vessel with two jackets for steam or water; its capacity to the top of the upper jacket is 76 liters; the 10 inch cylindrical section at the top encloses an equal volume which is the freeboard.
- (b) The entrainment separator, E-2, which is located on top of the evaporator combines a cyclone downdraft with ten inches of 1/2 inch by 1/2 inch Raschig rings.
- (c) The distillation column, E-3, is made up of nine feet of 4 inch pipe and six feet of 6 inch pipe; this column is packed with 7/8 inch by 7/8 inch Raschig rings.
- (d) The reboiler, E-4, is a conical vessel of 20 liters capacity. It has a single jacket for steam or water. A tantalum heater also extends into this vessel. The overflow point at which the recovered acid discharges to the catch tank represents a volume of 10 liters.
- (e) A number of additional vessels are shown schematically in Figure 3.41.

4.312 Instrumentation

Instruments are provided on the acid recovery unit to perform the following functions:

- (a) Record the level and specific gravity of the liquid in all the tanks and process vessels.

Instrumentation (continued)

- (b) Control the liquid level of the evaporator by operating a pneumatic valve supplying steam to the jackets.
- (c) Control the temperature of the vapors above the reboiler liquid by operating a pneumatic valve supplying steam to the jacket and the tantalum immersion heater.
- (d) Record temperature by means of thermocouples located at six points in the system.
- (e) Control the temperature of the reflux water to the fractionating column by operating a ~~balenoid~~ solenoid steam valve.
- (f) Record evaporator temperature and provide over-temperature alarm and emergency control in the event of malfunction of the liquid level controller or too high evaporator acid concentration.
- (g) Control the evaporator liquid temperature during final volume reduction.
- (h) Indicate water, steam, and air pressure to the jackets, spargers, and jets of the various vessels.

The 1/4 inch O.D. tubing originally run from the instruments to the evaporator and reboiler has been replaced by 1/4 inch IPS pipe to improve instrument performance. Thermowells drilled from bar stock were fabricated after welded units had failed.

4.32 Operation

4.321 Continuous Operation on Column Raffinates

The IA column raffinate, 2.2M in nitric acid and containing

Continuous Operation on Column Raffinates (continued)

the fission products removed from the uranium and plutonium, flows continuously to the evaporator during a run. The liquid in the evaporator is maintained at a constant level. If the system is at equilibrium, the vapors from the evaporator are 2.2M nitric acid and the liquid is approximately 8.5M nitric acid. Under these conditions the composition and weight of the vapor equals that of the entering raffinate. To reduce the possibility of oxidizing ruthenium to the volatile ruthenium tetroxide, a diluent stream of demineralized water equal to one half the volume of the IAW stream is pumped to the evaporator by a metering pump. The resulting equilibrium of acid and vapor phases produces approximately 1.5M nitric acid in the vapor and 6.5M nitric acid in the liquid.

The acid vapors after passing through the packing and cyclone of the entrainment separator are fed into the distillation column above the reboiler. The acid concentration of the reboiler is maintained constant by controlling the boiling temperature. The acid vapors boiling at a higher temperature tend to condense, concentrate and run down the column into the reboiler. Water vapor conversely tends to rise as steam to the top of the column and pass out through the condenser to the condensate tank. To reduce acid losses to the condensate additional reflux of demineralized water is pumped into the top of the column by a metering pump. The concentrated acid accumulating in the reboiler overflows to a catch tank and is held for disposition on the basis of analytical results.

4.322 Final Volume Reduction

Continuous evaporation of the raffinate reduces the entire

Final Volume Reduction (continued)

IAW volume to 76 liters. An additional five fold volume reduction is obtained by pumping water at eight gallons per hour to the evaporator, which is maintained at the boiling temperature of 6.5M acid. The remainder of the system functions in the same manner as before.

4.33 Results

4.331 Results of Cold Runs

Several preliminary runs made from dummy acid established the fact that the reboiler acid concentration could be controlled up to 65 percent. The IAW from two cold column runs (Figure 14) was evaporated in the acid recovery unit. The main results of interest were:

- (a) Because of the short duration of the runs volume reduction factors were limited to approximately five.
- (b) No water diluent was fed to the evaporator with the IAW, consequently the evaporator equilibrium nitric acid concentration was approximately 8M.
- (c) The final evaporator acidity in Run ISP-9 was greater than 9M because the average concentration of entering IAW was 0.3M higher than during the previous run.

Run ISP-9 was made with a 10 percent smaller reflux to distillate ratio and lower reboiler temperature control setting than Run ISP-8. The net result of these changes was the recovery of a larger volume of slightly more dilute acid accompanied by somewhat lower losses to the condensate.

4.332 Results of Hot Runs

Two hot runs have been made to date. IAW from ORNL level Run IHP-3 was fed to the acid recovery unit for several intermittent periods. Because of excessive boiling in the evaporator due to liquid level controller failure considerable activity entered both the acid and condensate streams. Peak values of  $2 \times 10^6$  and 400 beta cts/min/ml were determined in one set of flowing stream acid and condensate samples respectively. The composite acid reached a peak of  $6 \times 10^6$  beta cts/min/ml. The beta activity of the feed due to ruthenium was 6.3 percent. Analysis from the run of both flowing stream and composite samples yielded a range of ruthenium beta of 5-8 percent of total activity from six sets of samples including the final samples. This is an indication that the contamination was primarily due to entrainment of liquid rather than volatilization of ruthenium tetroxide out of the evaporator.

The entire IAW from Run IHP-5 was sent to the evaporator. The volume reduction was carried out as described in Section 3.42, namely continuous boildown at constant liquid level during the run, followed by a three-fold volume reduction at constant temperature. Figure 15 presents a summary of the data obtained. Points of note are:

- (a) During the run water was fed to the evaporator in a proportion of one part to two parts of IAW. During the run the temperature of the liquid in the evaporator remained at  $113^\circ\text{C}$  by virtue of the equilibrium conditions established.

Results of Hot Runs (continued)

- (b) A seven-fold volume reduction of IAW was achieved during the continuous boildown. A further three-fold volume reduction was carried out during the batchwise final boildown to yield a total IAW volume reduction factor of twenty-one.
- (c) The average beta decontamination factor from evaporator to recovered acid from both flowing stream and composite samples was  $4 \times 10^5$ .
- (d) A comparison of ruthenium concentration in the feed and the recovered acid indicated that ruthenium contamination in the recovered acid was due to entrainment and not volatilization.
- (e) The decontamination factor for the condensate was approximately  $10^6$ .
- (f) The average strength of the recovered acid from twenty-six flowing stream samples was 56 percent nitric acid.
- (g) The loss to the condensate from the overhead of the packed distillation tower was 1.9 percent.
- (h) The beta activity of the recovered acid was  $10^{-2}$  microcuries/ml; that of the condensate approximately  $10^{-4}$  microcuries/ml.
- (i) No control difficulties were encountered other than those due to oversized steam control valves which gave coarse control.
- (j) It was intended to maintain the acidity in the evaporator at 6.5M; however, the equilibrium temperature of the evaporator liquid and the analytical results showed that the acidity was close to 9M. It is assumed that the repacked entrainment separator furnished fractionation equivalent to one theoretical stage. This was confirmed by the



Results of Hot Runs (continued)

- (j) temperature differences between the acid vapors entering the column and the liquid in evaporator.

4.34 Program

IAW from hot runs will be fed to the acid recovery unit. Between hot runs the system will be operated on cold acid. Points to be investigated are:

- (1) Decontamination as a function of volume reduction factor.
- (2) The effect of varied percent water dilution of the IAW on decontamination.
- (3) The effect of varied column reflux ratio upon nitric acid losses.
- (4) Corrosion of a number of test specimens placed throughout the system.
- (5) The possibility of recycling a portion of the condensate collected.

4.4 Solvent Recovery

Solvent used in the pilot plant is now recovered by a batch process employing remotely operated equipment in which phases are distinguished by conductivity probes. For details of this equipment see Figure 7. Washing is done in 200 gallon batches with one 0.2 volume wash with 0.1M sodium carbonate, followed by two 0.1 volume demineralized water washes. Contact time for the carbonate wash is one hour, and a final settling time of 20 minutes is allowed. The above procedure has been effective in reducing uranium content from about 0.2 g/l to less than 0.001 g/l. ICW beta activity from ORNL level runs has been reduced from  $10^4$  c/min/ml to 50-120 c/min/ml. New makeup solvent is treated in the

Solvent Recovery (continued)

same manner. While this system has reduced uranium and beta contamination to satisfactory levels, a continuous system would be more applicable to a full scale plant. Equipment flowsheets of two continuous systems under consideration are given in Figures 8 and 9.

In alternate No. 1 the combined ICW and IEW streams are fed into the bottom of a pulse column in which sodium carbonate solution enters the middle to wash the solvent of activity and organic impurities. Water enters the top of the column to free the solvent of carbonate.

Alternate No. 2 treats the ICW in a preliminary wash column with sodium carbonate to reduce its activity to the level of IEW. The washed ICW and unwashed IEW enter a pulse column operated in the same fashion as in alternate No. 1. The exit carbonate stream from the second column is used to contact the ICW in the preliminary column.

Preliminary data indicate a more favorable distribution coefficient for cleanup with a higher beta activity than with lower activity in the solvent. Investigation of a continuous dielectric constant instrument to control the solvent TBP concentration has been initiated. In both alternates No. 1 and No. 2 such a controller would add Amsco to the washed solvent or TBP to untreated solvent as required.

During the next quarter equipment for the continuous recovery of solvent will be installed in the Pilot Plant.

4.5 Product Evaporation

The IBsP, IIBsP, ICU and IEU product streams are evaporated continuously

Product Evaporation (continued)

to reduce the volume of the solutions for crossover to the second cycles and for final product solution withdrawal.

IBsP Evaporation

The plutonium product from the first cycle flows continuously into a 60 gallon evaporator which is maintained at constant liquid level. During each run the volume of the IBsP is reduced by a factor of five and the acidity is increased from 1.25M to 6.0M. Evaporation losses have been below 0.1%.

ICU Evaporation

The first cycle uranium product is jettied every few hours to an evaporator equipped with a steam coil and jacket and a packed entrainment separator. The solution is concentrated to about 2M in uranium. Condenser capacity has limited evaporation to 30 gals/hr at which rate uranium losses have been below 0.01 percent.

IEU Evaporation

At present the second cycle uranium product is continuously evaporated as it is received. The evaporator is equipped with a steam coil and jacket and a packed entrainment separator. Boildown rates of 15 gals/hr are being used with uranium losses below 0.01 percent. The final product is 2M with respect to uranium. During the next quarter a new evaporator designed by Blaw-Knox will be installed to evaporate the IEU stream. This evaporator will be an exact replica

IEU Evaporation (continued)

of evaporators to be used at the Savannah River Project.

IIBsP Evaporation

The second plutonium cycle product is jetted in batches to a jacketed evaporator in which it is boiled down continuously at an average rate of 2 gals/hr to a volume of one liter. Vapors pass through a baffled pipe to the condenser. Plutonium losses to the condensate have been below 0.1 percent.

. 4.6 Purex Wastes

The waste streams of the Purex process in the pilot plant originate from two sources: (1) The primary waste streams, i.e. the actual process streams which contain insignificant amounts of uranium and plutonium, and (2) The secondary waste streams which result from the recovery of the solvent and nitric acid from the primary waste streams.

The volumes and approximate compositions of the primary wastes are summarized in Table 16 and those for the secondary wastes in Table 17. The volumes given are on the basis of 100 volumes of feed. The fission product and plutonium counts reported are based on pilot plant runs using 100 g/t feed material. The stream designations are shown in the chemical flowsheet (Figure 12).

A pilot plant program to develop and demonstrate a flowsheet for waste treatment is in progress. This program is directed toward a substantial decrease in the volume of hot waste to be stored and the recycle of the water recovered.

[REDACTED]

4.7 Program

Dissolving

Final design and installation of fumeless dissolving equipment will be completed during the summer.

Solvent Extraction

After the conclusion of a short series of scouting runs at Hanford level, an engineering program is planned in which uranium, plutonium and fission product separation in the Purex process will be investigated as functions of pulse column variables such as pulse amplitude, pulse frequency, pulse shape, plate spacing, plate hole diameter and throughput. These runs will be made using 100 g/t feed material from the ORNL pile.

Nitric Acid Recovery

Development of alternate operational procedures and corrosion testing will be continued in the nitric acid recovery system.

Solvent Recovery System

Further development and installation of a continuous solvent recovery system is scheduled for the summer.

5.0 SCRUP Program - E. C. Stewart

The Laboratory has agreed to a proposal by the Commission whereby the irradiated fuel rods from the Chalk River pile would be processed for plutonium

SCRUP Program (continued)

in the ORNL Metal Recovery Plant. This new program entails the processing of a large number of irradiated rods now in the Chalk River Canal in addition to the processing of rods to be discharged during the next several years. The new program has been designated as the SCRUP Program (Separation of Chalk River Uranium and Plutonium). The Metal Recovery Program will be deferred until January, 1952 while the backlog of Chalk River material is being processed; at that time the two programs may be run concurrently. Capital additions required for the new program will cost about \$91,000 and the first hot runs using Chalk River material are scheduled for August, 1951.

5.1 Equipment Alterations and Modifications

The SCRUP Program will require only minor alterations to the equipment already planned for the Waste Metal Recovery Program. Major equipment additions will be required including three carriers, storage basin, dissolver with off-gas facilities and additional plutonium handling equipment.

5.2 Shipping Schedule

The fuel rods are to be sheared in twelve-inch lengths and 100 lengths will be shipped in each carrier. A railway express car will be leased full time for the shipments, which are planned to begin about July 1, 1951 and which will continue at the rate of one per month for the duration of the program.


### 5.3 Process Flowsheet

It is currently planned to employ the Purex Flowsheet No. 1 for the SCRUP Program. The plutonium will be processed through one cycle of solvent extraction followed by an ion exchange concentration step, a second solvent cycle and removed from the system to isolation in an ion exchange cartridge bed. The uranium will be partially recycled to provide column equilibrium conditions prior to the starting of the feed to the columns and subsequent to feed run-out time. However, the bulk of the uranium will be stored in the waste tank farm for further processing during the Metal Recovery Program at a later date.

The detailed process conditions will be governed largely by the conditions of criticality and batching. Storage of process raffinates is contemplated.

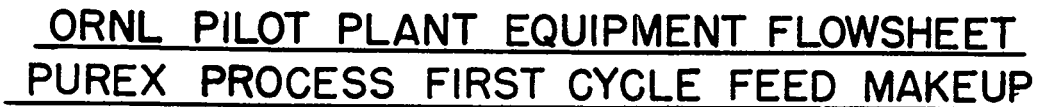
### 5.4 Program Status

The original plant is estimated to be 70 percent complete. Close coordination of the construction may incorporate many of the direct alterations required by the SCRUP Program, thus reducing the expenditures. The additions and alterations are expected to extend the construction approximately five weeks. Initial hot runs using Chalk River fuel are scheduled for August, 1951.

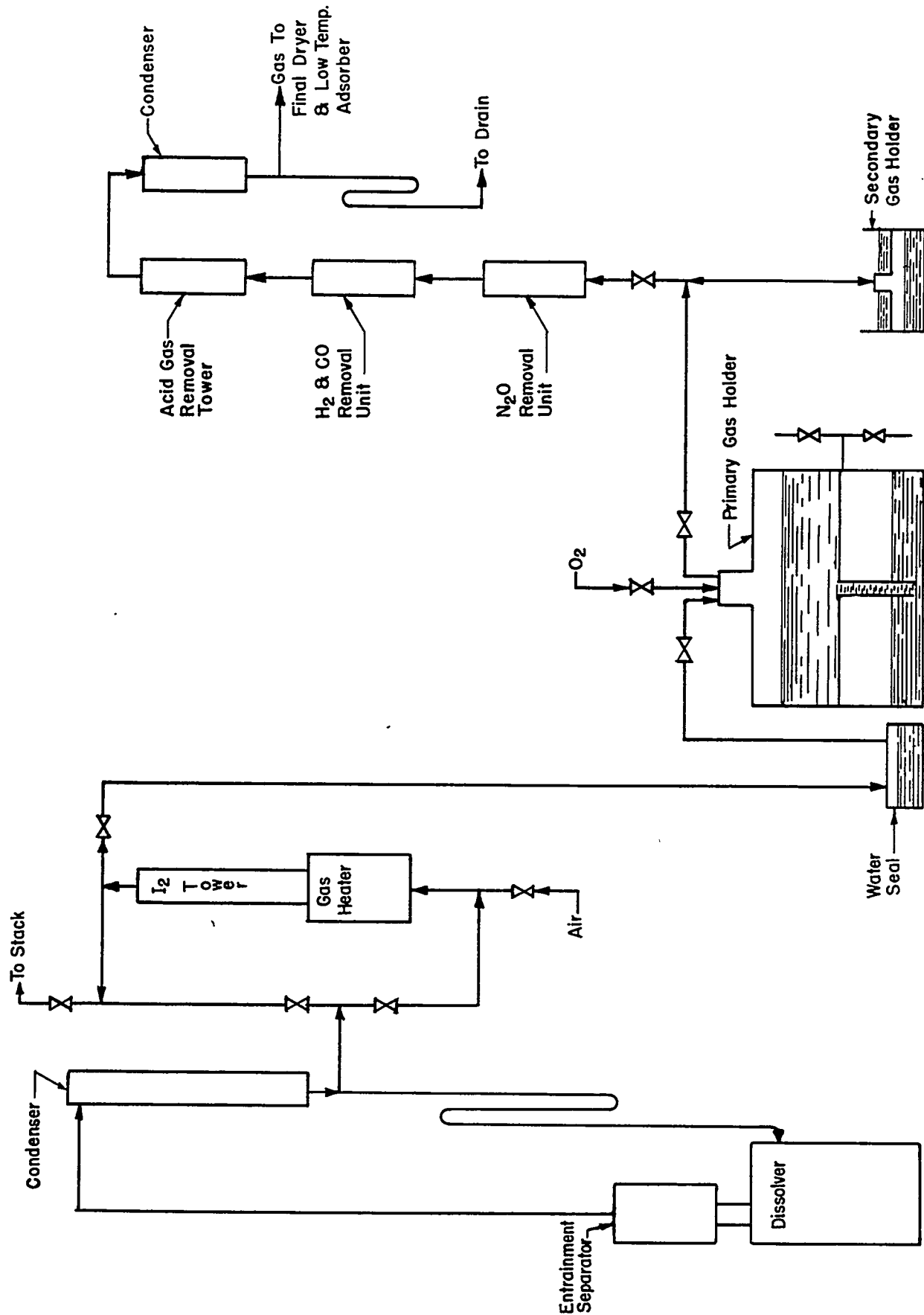


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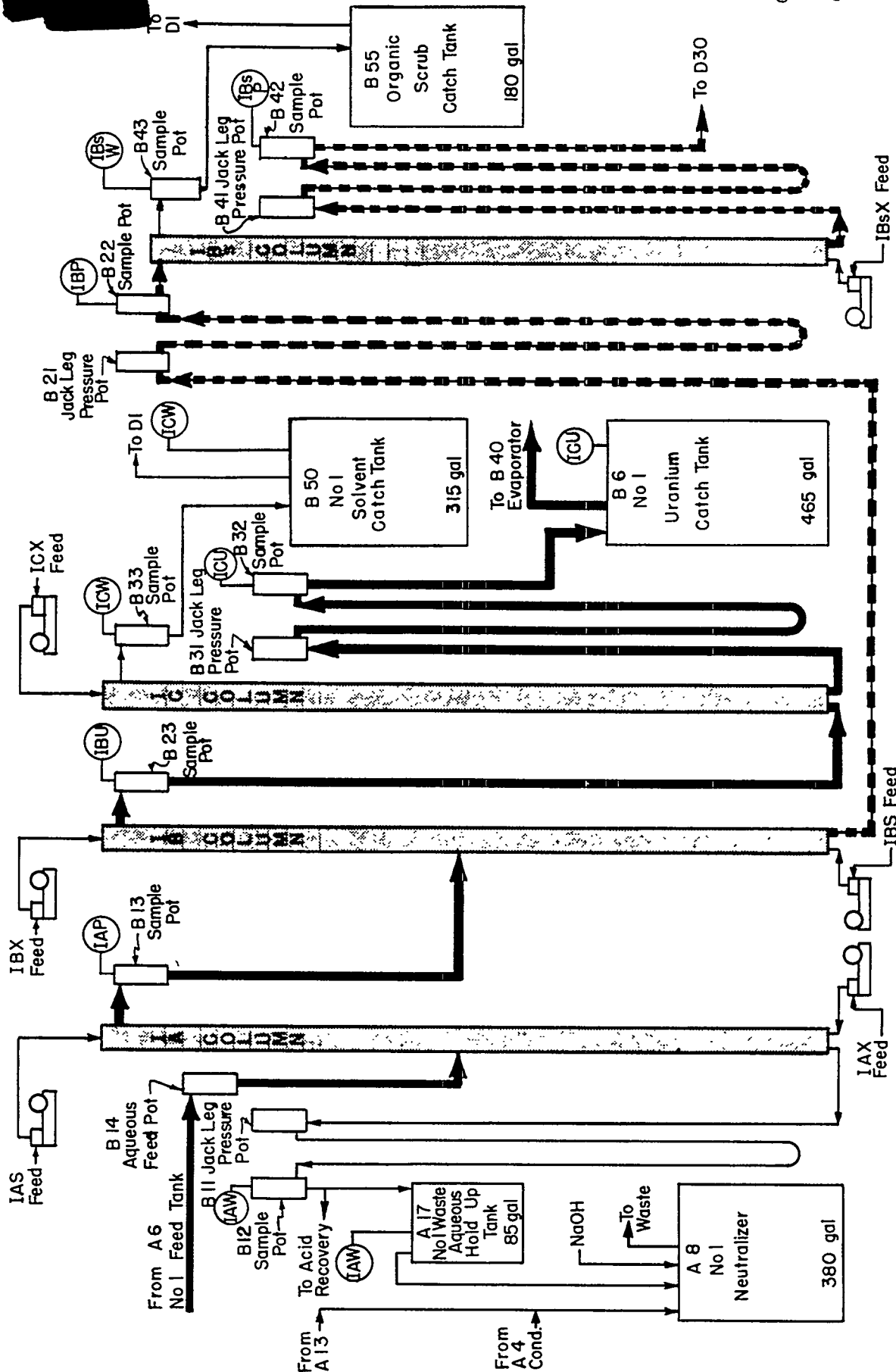




**ORNL PILOT PLANT EQUIPMENT FLOWSHEET  
PROPOSED FUMELESS DISSOLVING SYSTEM**

5-14-51

Figure 3



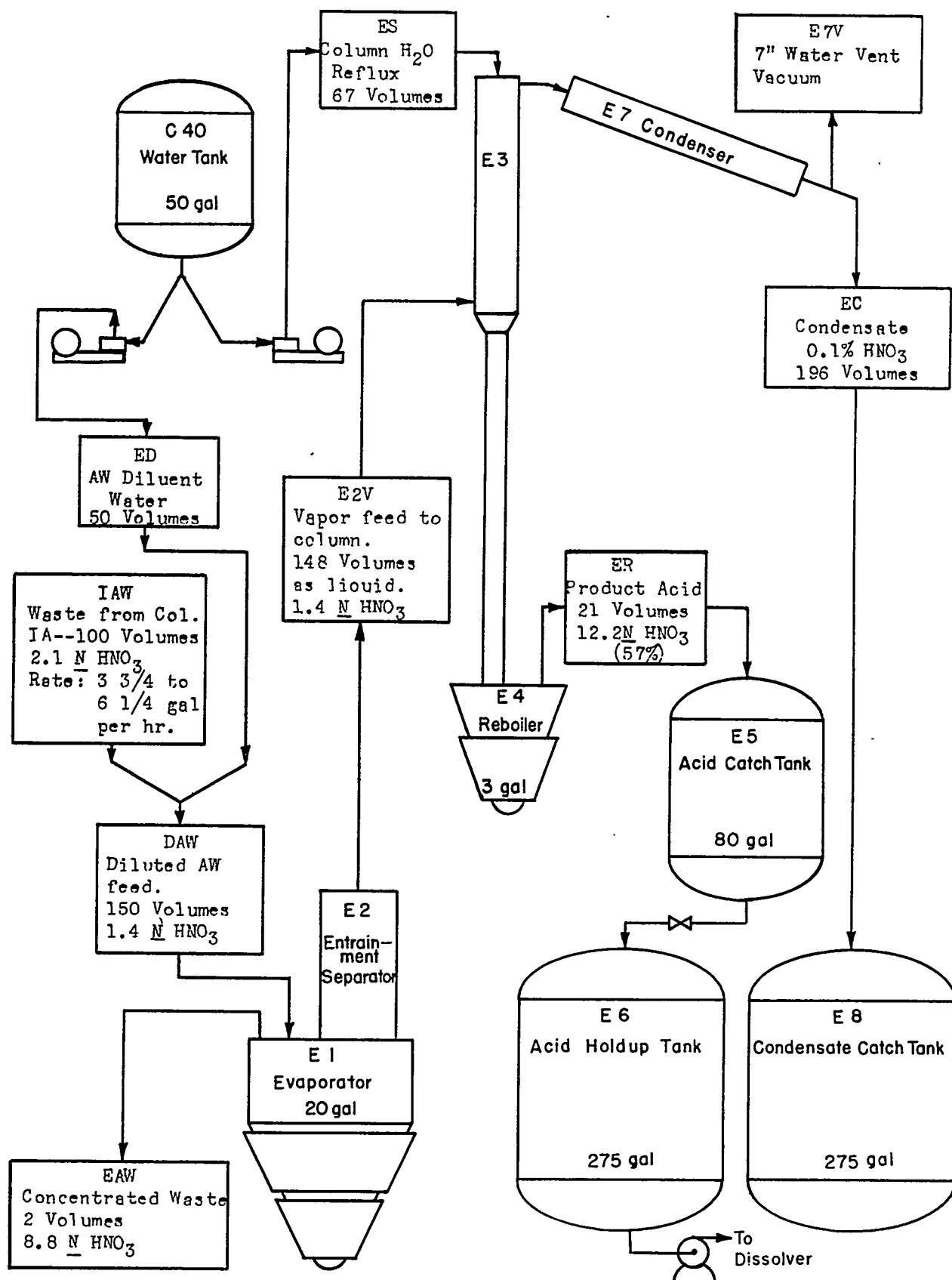
ORNL PILOT PLANT EQUIPMENT FLOWSHEET  
PUREX PROCESS FIRST URANIUM & PLUTONIUM CYCLE

[illegible]

[REDACTED]

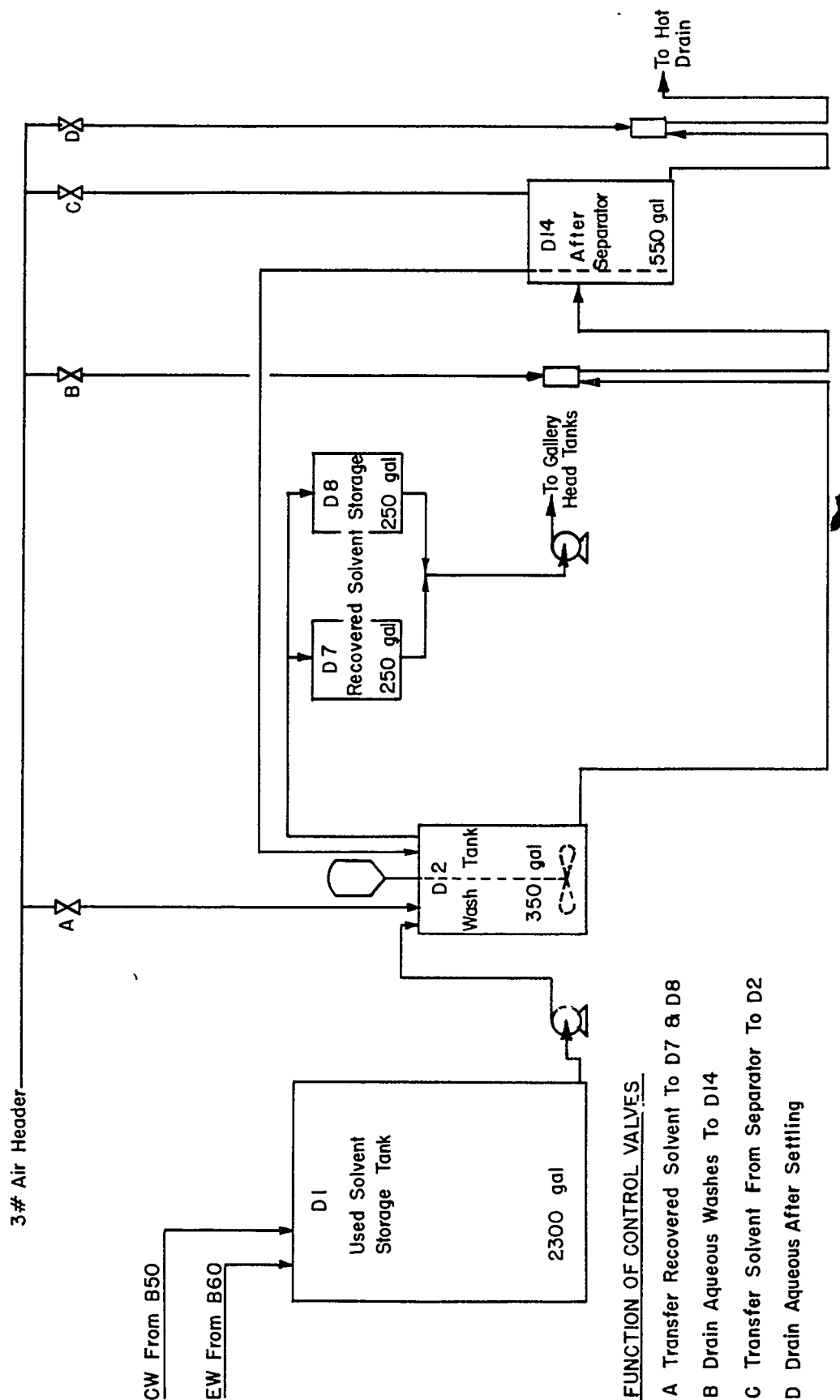
Figure 5





**ORNL PILOT PLANT EQUIPMENT FLOWSHEET**  
**PUREX IAW ACID RECOVERY UNIT**

Figure 7



FUNCTION OF CONTROL VALVES

- A Transfer Recovered Solvent To D7 & D8
- B Drain Aqueous Washes To D14
- C Transfer Solvent From Separator To D2
- D Drain Aqueous After Settling

ORNL PILOT PLANT EQUIPMENT FLOWSHEET

PRESENT SOLVENT RECOVERY SYSTEM

5-14-51

Figure 8

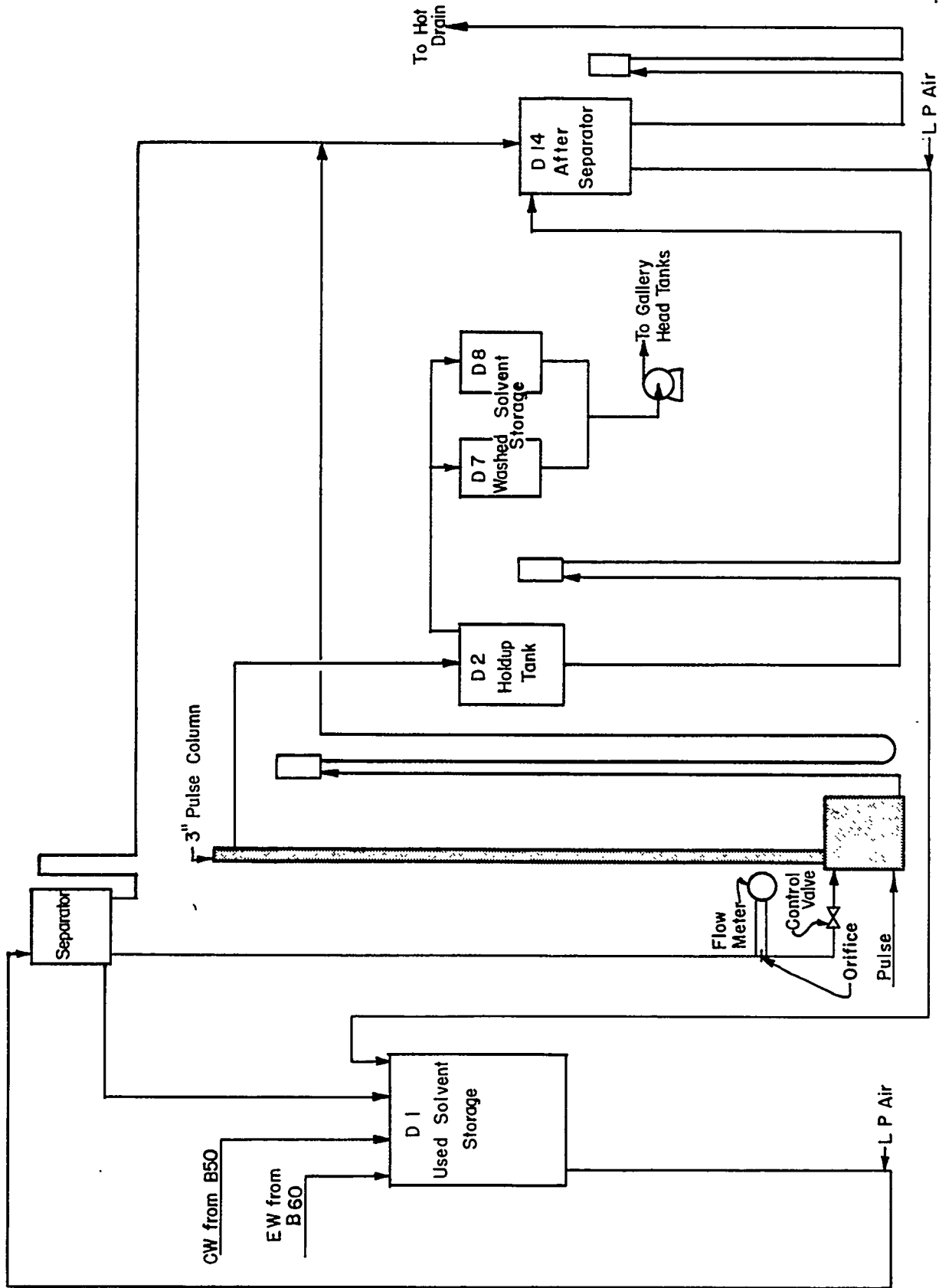
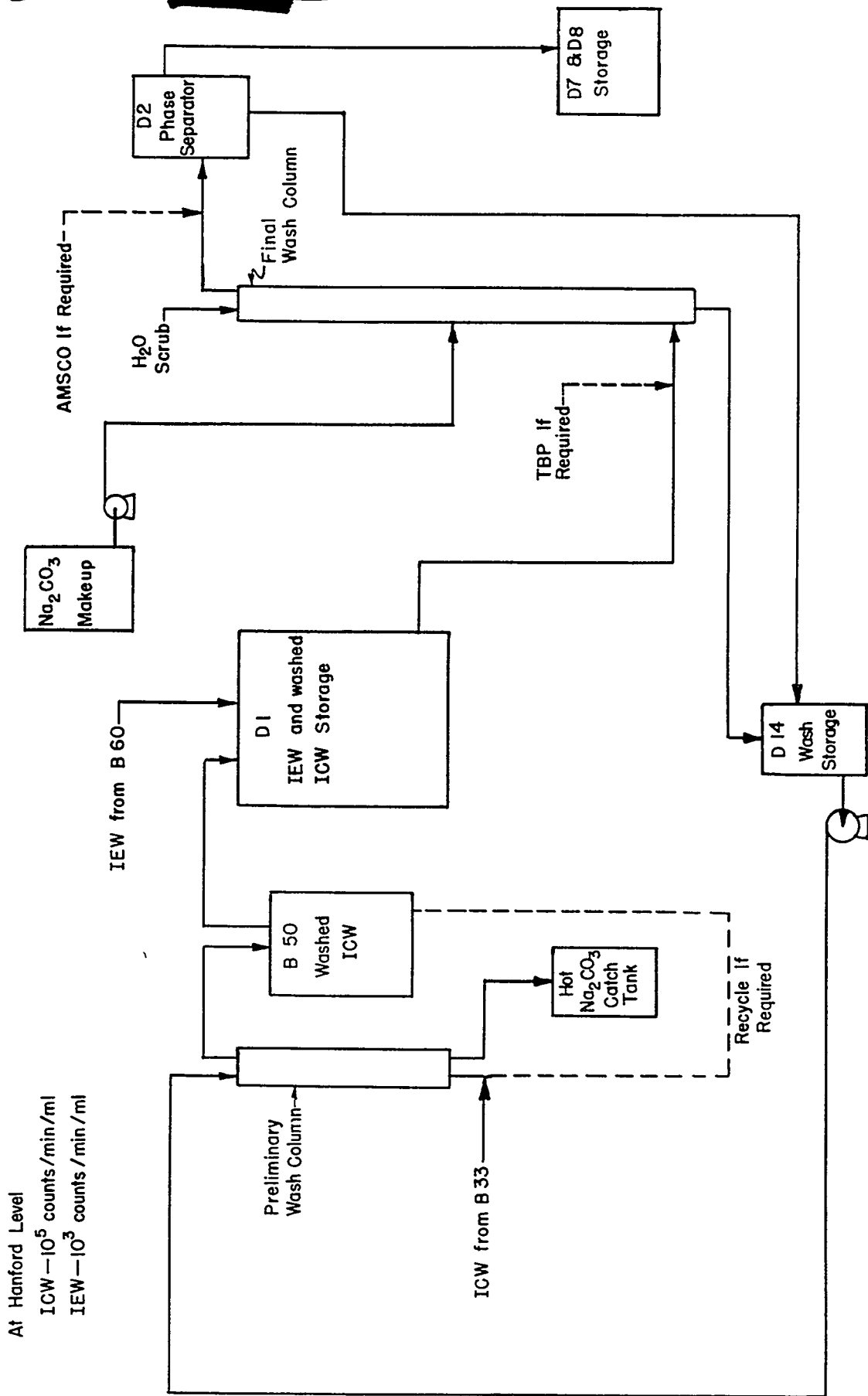


Figure 9

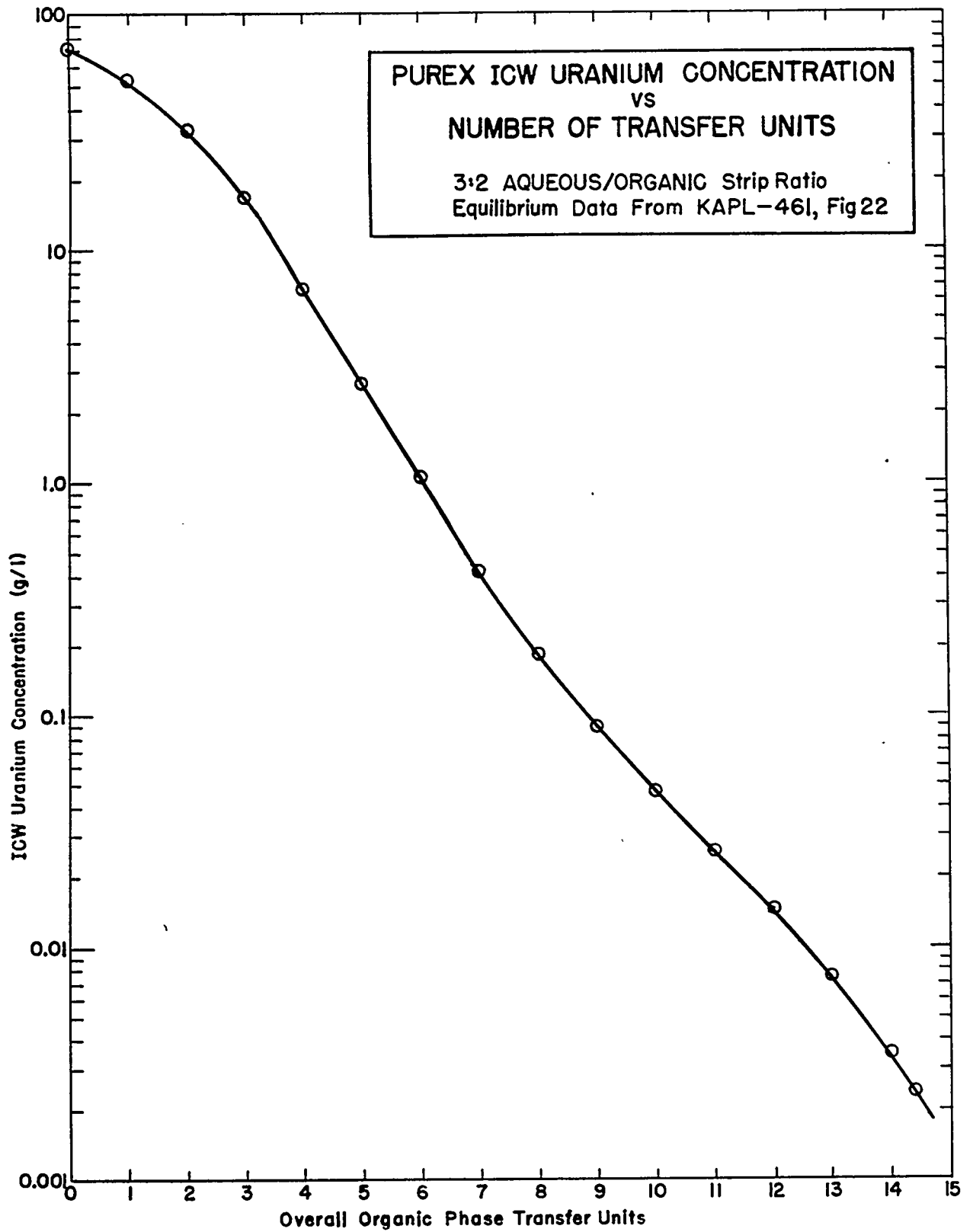


At Hanford Level

ICW — 10<sup>5</sup> counts/min/ml

IEW — 10<sup>3</sup> counts/min/ml





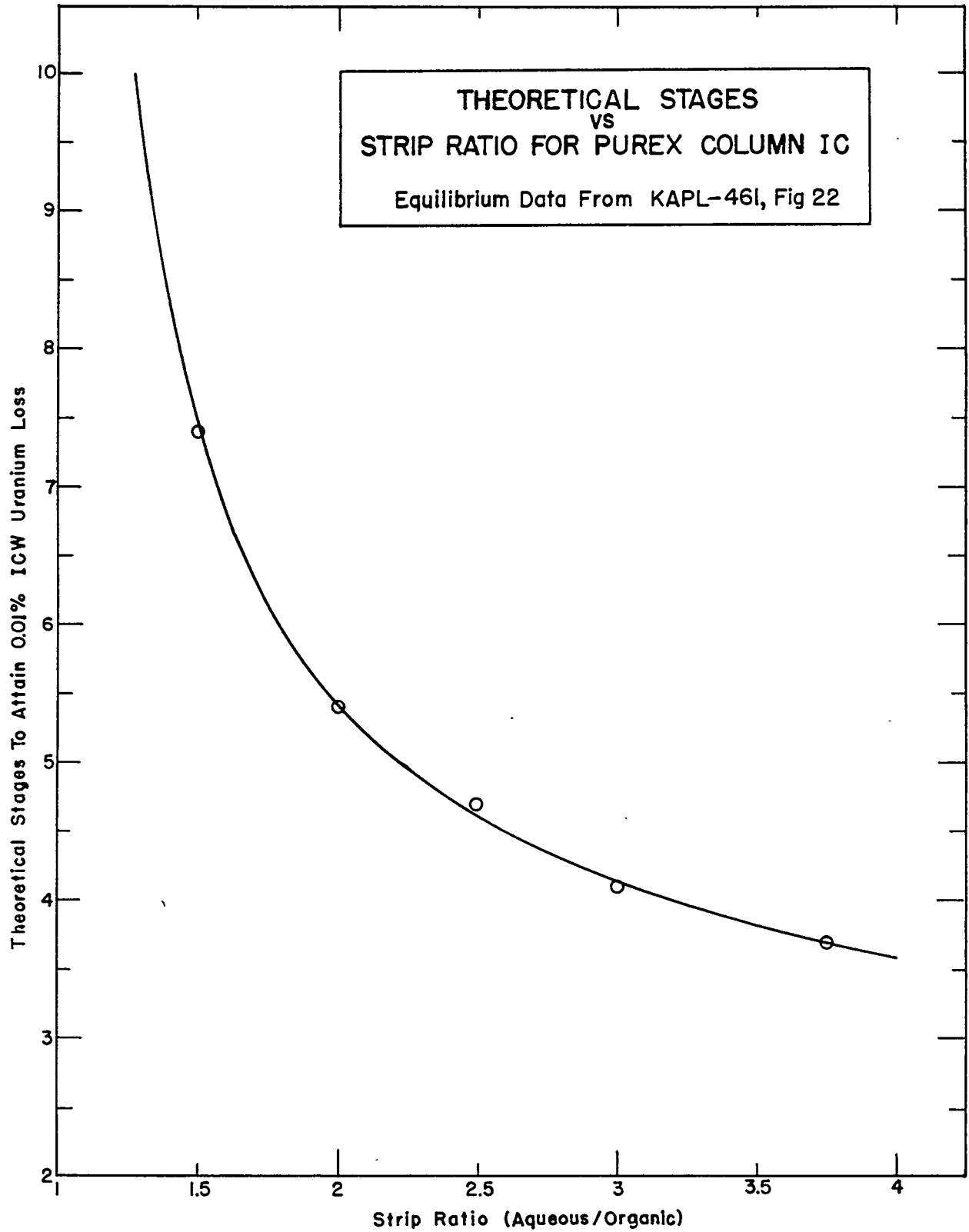


Figure 12  
PUREX PROCESS - FLOWSHEET NO. 1

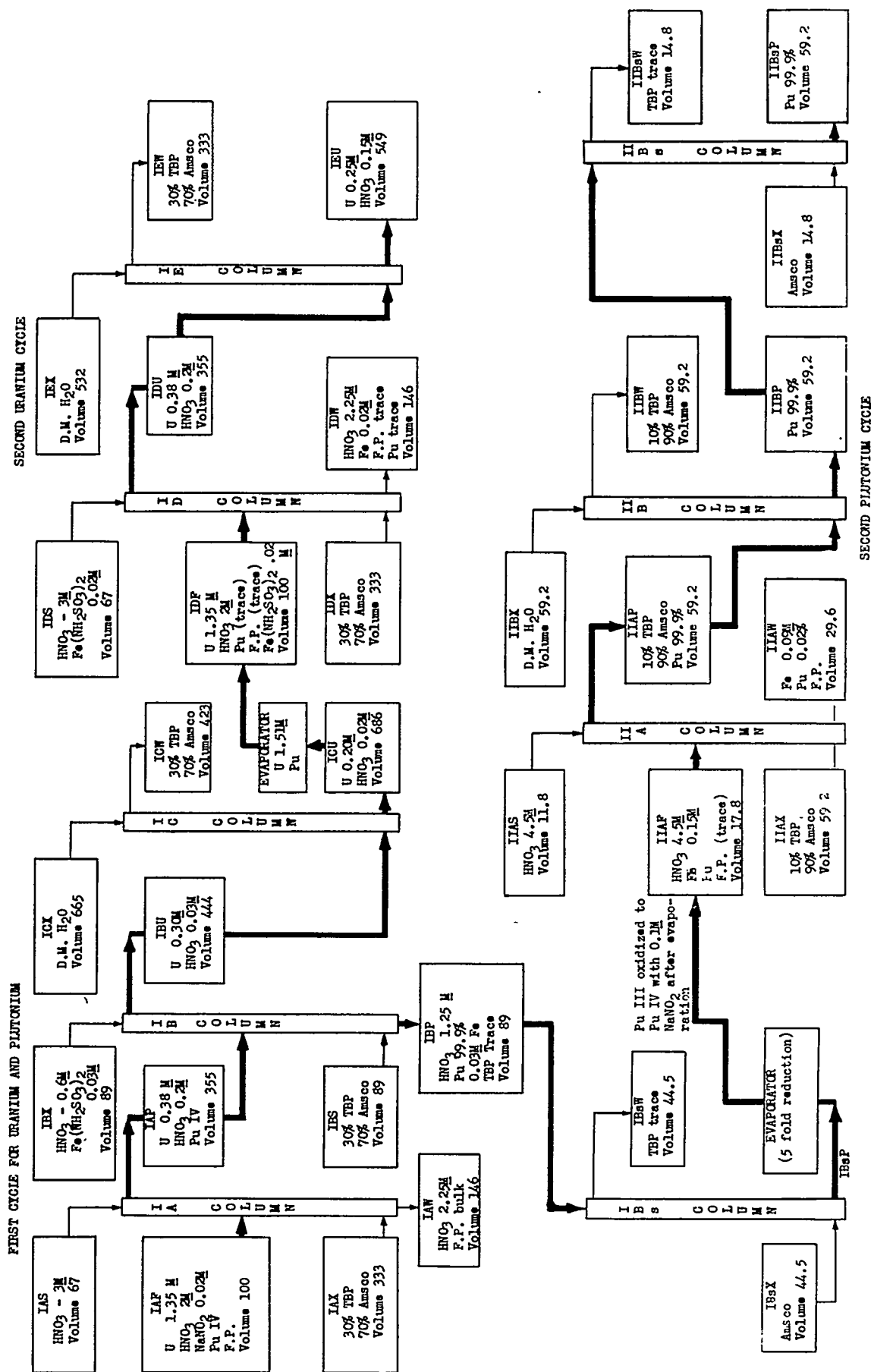


Figure 13  
PUREX PROCESS FLOWSHEET NO. 2

Dwg. 11619

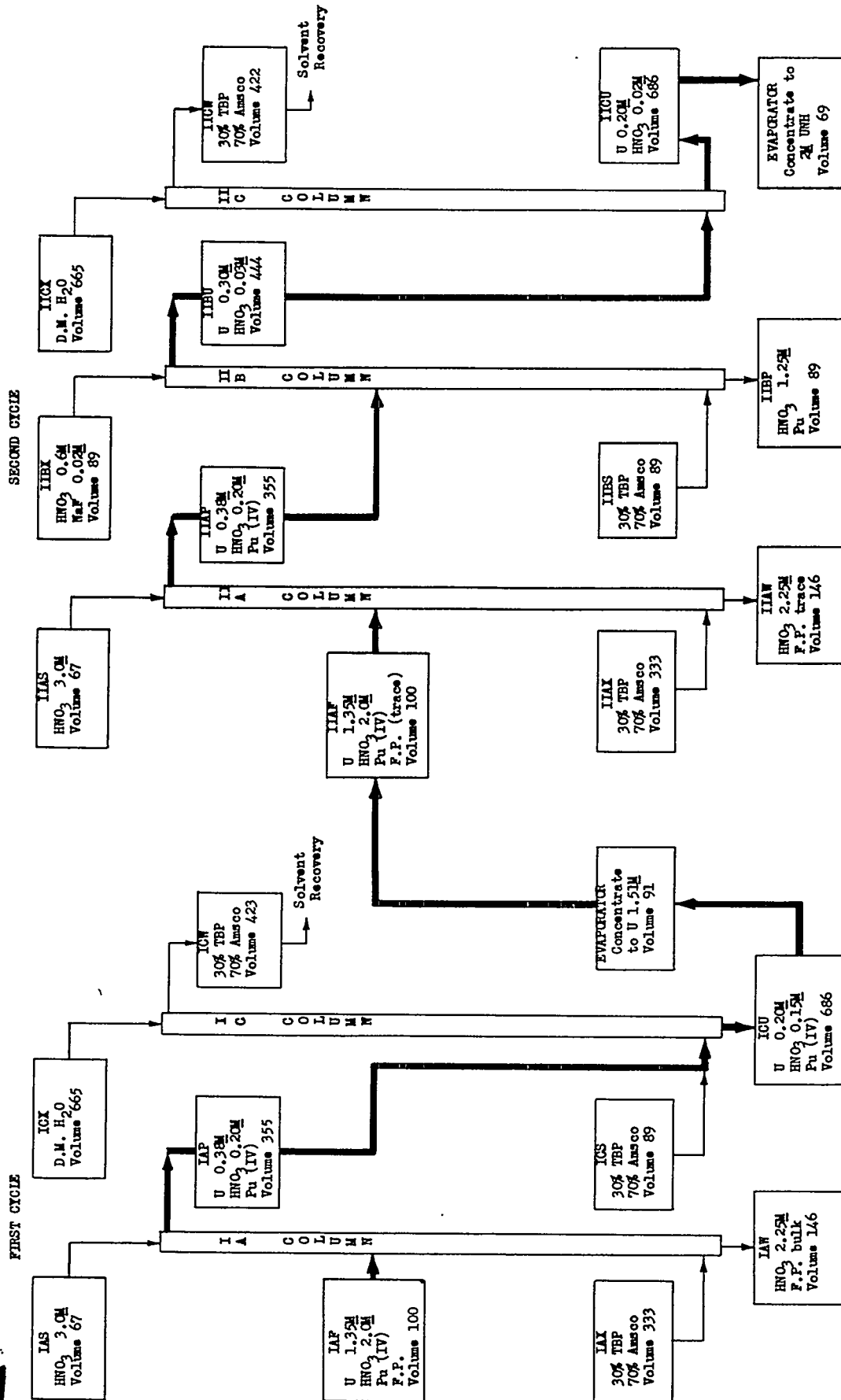


Table 1  
PUREX COLD DISSOLVINGS

Run No.	ISP-3a	ISP-3b	ISP-10a	ISP-10b	ISP-11a	ISP-11b
Initial Metal	143 X slugs	143 X slugs	143 X slugs	143 X slugs	101 W slugs 63 X slugs	~167 X slugs
Jacket Removal Loss	-	-	-	-	0.008 %	0.023 %
% HNO <sub>3</sub> charged	55	55	50	45	55	55
Initial molar ratio HNO <sub>3</sub> /U (allowing for heel)	5.5	5.5	5.0	4.0	5.0	5.0
Dissolved	61 X slugs	72 X slugs	84 X slugs	80 X slugs	~55 X slugs	~81 X slugs
% Heel at End	136	100	71	79	340	107
Product sp. gr. at 100°C	1.51	1.56	1.63	1.70	1.52	1.62
Product U (g/l)	346	407	432	488	355	456
Product HNO <sub>3</sub> (N)	1.13	0.43	1.95	1.28	0.75	0.47
Product Volume (l)	206	204	230	193	183	210
Mols HNO <sub>3</sub> charged	1970	1970	1785	1450	1790	1790
Mols NO <sub>3</sub> associated with product	606	698	835	790	546	804
Mols free HNO <sub>3</sub> in product	233	88	449	247	137	99
Mols HNO <sub>3</sub> consumed per Mol U dissolved	5.2	5.4	3.2	3.1	6.0	4.2
% HNO <sub>3</sub> utilized	42.6	39.8	71.9	71.5	38.1	50.4

ISP-3a - Updraft condenser, slug chute not tight  
 ISP-3b - Updraft condenser, slug chute not tight, condenser jacket leaked into dissolver  
 ISP-10a - Down draft condenser, slug chute not tight, A2 ring spray on  
 ISP-10b - Down draft condenser, slug chute not tight, A2 ring spray on  
 ISP-11a - Down draft condenser, slug chute fairly tight, jackets only partially removed  
 ISP-11b - Down draft condenser, slug chute fairly tight

Table 2  
PUREX HOT DISSOLVINGS

Run No.	IHP-1a	IHP-1b	IHP-2a	IHP-2b	IHP-3a	IHP-3b	IHP-4a	IHP-4b	IHP-5a	IHP-5b
No. of X-Slugs Charged	130	-	129	-	129	-	216	-	156	-
Moles of U Initially	1065	655	1060	699	1000	650	1058	659	1049	697
Jacket Removal Loss %	<0.01-U	-	<0.01-U	-	0.01-U	-	0.01-U	-	0.11-U, 0.04 Fu	-
Initial Molar Ratio										
HNO <sub>3</sub> /U (allowing for desired heel)	5.1	5.5	5.0	5.0	5.5	5.6	5.1	5.5	5.1	5.2
Mols U dissolved	410	295	316	329	350	336	399	374	352	377
% Heel at End	160	122	194	112	186	93	165	76	198	85
Product Sp.G. before dilution at 100°C	1.73	1.73	1.69	1.70	1.72	1.72	1.72	1.73	1.73	1.76
Product U(g/l) after dilution	444	448	413	425	412	398	416	414	391	412
Product HNO <sub>3</sub> (N) after dilution	0.06	0.56	0.25	0.73	0.18	0.44	0.21	0.46	0.63	0.26
Product Volume (l) after dilution	220	157	208	184	202	201	228	215	214	218
Mols NO <sub>3</sub> associated with product	820	590	722	658	700	672	797	748	704	754
Mols free HNO <sub>3</sub> in product	13	88	52	134	36	88	48	99	135	57
Mols HNO <sub>3</sub> consumed/Mol U dissolved	4.4	5.8	4.8	5.0	5.1	5.2	4.4	4.6	4.7	4.6
% HNO <sub>3</sub> Utilized	46.1	37.5	43.2	44.2	40.2	41.5	44.0	41.3	46.6	45.1
Dissolution time, hours	12	10	8	9	11	14	10	9	7	15

1800 mols 55% HNO<sub>3</sub> charged, tight slug chute, downdraft condenser, no water spray during dissolution for all dissolvings.

Table 3  
PUREX FLOWSHEET #1 COLD FIRST CYCLE PACKED COLUMN RUNS

Run No.	IS-1	IS-2	IS-3	IS-4
Kg. Metal Per Day	50	50	37.5	25
Duration (Hours)	49	46	46	55
Gals/hr/ft. <sup>2</sup> (both phases)				
IA Extraction	160	160	120	80
IA Scrub	140	140	105	70
IB Partioning	150	150	110	75
IB Scrub	60	60	45	30
IBs	-	-	60	40
IC	210	250	160	200
% U Losses (Flowing Stream Average)				
Column IA	0.005	0.001	<0.001	<0.001
Column IB	0.001	0.0002	0.004	0.063
Column IC	5.00	1.95	2.55	0.070
% U Losses (Composite)				
Column IA	0.007	0.055	< 0.001	<0.001
Column IB	0.001	0.005	0.002	2.92
Column IC	3.98	1.31	1.76	0.074
CX/BU Ratio	3/2	2/1	3/2	15/4
% Material Balance (Flowing Stream)				
U	96.5	95.5	110.3	96.1
HNO <sub>3</sub>	97.6	114.9	102.3	98.4
% Material Balance (Composite)				
U	96.1	106.9	103.8	59.8
HNO <sub>3</sub>	95.3	105.3	102.1	69.6
Solvent	102.5	112.3	88.2	124.7

(Table continued on next page)

Table 3

PUREX FLOWSHEET #1 COLD FIRST CYCLE PACKED COLUMN RUNS (continued)

<u>Section</u>	<u>Contacting Height (ft.)</u>	<u>Diameter (in. IPS)</u>
IA Extraction	20	3
IA Scrub	20	3
IB Partioning	28	3
IB Scrub	13	3
IBs	15	2
IC	39	4

All columns packed with 1/4" x 3/8" Raschig Rings

Flowsheet #1

<u>Stream</u>	<u>Composition</u>	<u>Relative Volume</u>
IAF	1.35M UNH, 2.0N HNO <sub>3</sub> , 0.02M NaNO <sub>2</sub>	100
IAS	3.0N HNO <sub>3</sub>	67
IAX	30% Tributyl Phosphate in Amsco 123-15	333
IBS	30% Tributyl Phosphate in Amsco 123-15	89
IBX	0.03M Fe(NH <sub>2</sub> SO <sub>3</sub> ) <sub>2</sub> - 0.6N HNO <sub>3</sub>	89
IBsX	Amsco 123-15	45
ICX	Demineralized Water	Varied with CX/BU strip ratio - 665 for 3/2 ratio



PUREX FLOWSHEET #1 COLD SECOND U CYCLE PACKED COLUMN RUNS\*

Run IIS-1 (combined with IIS-1a), 54 hour duration, 37.5  
Kg metal/day, 3/2 EX:DU strip ratio

Gals/hr/ft. <sup>2</sup> (both phases)	
ID Extraction	120
ID Scrub	105
IE	145
% U Losses (flowing stream)	
Column ID	0.005
Column IE	0.24
% U Losses (composite)	
Column ID	0.020
Column IE	0.22
% Material Balance (flowing stream)	
U	95.6
HNO <sub>3</sub>	101.5
% Material Balance (composite)	
U	64.5
HNO <sub>3</sub>	96.7
Solvent	87.4

<u>Section</u>	<u>Contacting Height (ft.)</u>	<u>Dia. (" IPS)</u>
ID Extraction	19	3
ID Scrub	20	3
IE	37	4

Flowsheet #1

<u>Stream</u>	<u>Composition</u>	<u>Relative Volume</u>
IDF	1.35M UNH, 2.0N HNO <sub>3</sub>	100
IDS	3.0N HNO <sub>3</sub>	67
IDX	30% Tributyl Phosphate in Amsco 123-15.	333
IEX	Demineralized Water	532

\*Run IIS-1a was a continuation of Run IIS-1 employing heated water strip. Since the IEW losses showed no change, the two runs were calculated together.

% U Losses (Composite)													
	Column IA	0.005	0.023	0.46	0.005	2.52	0.058	0.049	0.025	0.036	0.101	0.059	0.24
Column IB						0.003	0.060	0.004	0.0001	0.068	6.42	0.057	0.006
Column IC	3.60	2.17	1.13	0.008	2.06			0.042	0.012	0.14	3.17	0.050	
% Material Balance (Flowing stream)													
	U	91.0	86.8	89.4	91.3	97.6	105.0	110.0	102.6	94.7	89.7	86.6	108.6
HNO <sub>3</sub>	106.7	95.0	109.6	104.5	116.6	104.3	107.3	107.3	98.5	94.7	127.3	101.6	104.1
% Material Balance (Comp.)													
	U	92.0	88.1	83.2	92.9	93.0	96.2	96.2	93.8	93.7	83.3	85.9	111.1
	HNO <sub>3</sub>	104.1		79.7	104.6	107.6	102.5	105.8	109.7	97.1	129.6	102.4	104.1
	Solvent	89.2	98.1	106.1	106.1	96.1	98.7	85.0	72.0	100.1	103.1	132.0	80.6

Section	Diameter (in IPS)	Pulse Frequency (Cycles/min)
IA Extraction	2	58
IA Scrub	3	58
IB Partitioning	2.5	58
IB Scrub	2	58
IBS	2	Packed Column
IC	4	73

### FLOWSHEET #1

Stream	Composition	Relative Volume
IAF	1.35 M UNH <sub>3</sub> , 2.0 N HNO <sub>3</sub> , 0.02 M NaNO <sub>2</sub>	100
IAS	3.0 N HNO <sub>3</sub>	67
IAX	30% Tributyl Phosphate in Amsco 123-15	333
IBS	30% Tributyl Phosphate in Amsco 123-15	89
IBX	0.03 M Fe(NH <sub>2</sub> SO <sub>3</sub> ) <sub>2</sub> - 0.6 N HNO <sub>3</sub>	89
IBSX	Amsco 123-15	45
ICX	Deminerlized Water	665

ALL plates on 2" spacing, 1/8" diameter holes, 23% free area/plate.  
IAX and IBS streams pulsed. IC column pulsed directly.

IBS column packed with 1/4" X 3/8" Raschig Rings.

Table 5

## PUREX FLOWSHEET #1 COLD FIRST CYCLE PULSED COLUMN RUNS

Run No.	ISP-1	ISP-2	ISP-3	ISP-4	ISP-5	ISP-6	ISP-7	ISP-8	ISP-9	ISP-10	ISP-11	ISP-12
Kg metal/day	150	150	150	125	125	125	150	150	150	125	125	125
Duration hours	24	28	22	24	26	24	24	24	24	31	33	29
Gals/hr/ft <sup>2</sup> (both phases)	1095 410 780 375 235 615	1095 410 780 375 235 615	1095 410 780 375 235 615	910 340 650 310 -- 510	910 340 650 310 -- 510	910 340 650 310 -- 510	1095 410 780 375 -- 615	1095 410 780 375 -- 615	1095 410 780 375 235 615	910 340 650 310 195 510	910 340 650 310 195 510	910 340 650 310 195 510
Contacting Height (ft)  1A Extraction 1A Scrub 1B Partitioning 1B Scrub 1BS 1C	8.5 8 8 8 15 12	8.5 8 8 8 15 12	8.5 8 8 8 15 12	8.5 8 8 8 -- 12	8.5 8 8 8 -- 12	8.5 8 8 8 -- 12	7.5 8 8 8 -- 12	7.5 8 8 8 -- 12	7.5 8 8 8 15 12	7.5 9 11.5 6 15 12	7.5 12.5 11.5 6 15 12	7.5 12.5 11.5 6 15 12
Pulse Amplitude In Column (in.)  1A Extraction 1A Scrub 1B Partitioning 1B Scrub 1C	0.77 0.34 0.53 0.77 0.42	0.92 0.41 0.64 0.92 0.49	1.08 0.48 0.64 0.92 0.56	1.08 0.48 0.64 0.92 0.56	0.92 0.41 0.53 0.77 0.49	0.77 0.34 0.53 0.77 0.56	0.62 0.27 0.43 0.62 0.56	0.77 0.34 0.53 0.77 0.56	0.92 0.41 0.43 0.62 0.56	0.92 0.41 0.53 0.77 0.49	0.77 0.34 0.53 0.77 0.56	0.92 0.41 0.58 0.85 0.56
% V Losses (flowing stream)												
Column 1A	0.002	0.018	0.009	0.006	0.001	0.051	0.043	0.010	0.025	0.039	0.070	0.031
Column 1B	0.002	0.013	0.10	0.0008	0.0001	0.041	0.005	0.0045	0.029	0.014	0.014	0.007
Column 1C	3.32	2.02	1.15	0.020	2.61	0.36	0.042	0.011	0.033	2.70	0.050	0.045

Material Balance (Flowing Stream) %								
U		102.6	99.3	89.7	-	103.9	103.3	91.4
HNO <sub>3</sub>		91.1	96.8	92.6	-	102.3	102.0	103.4
Material Balance (Composite) %								
U		99.4	106.1	96.2	-	95.6	102.6	93.8
HNO <sub>3</sub>		94.2	99.9	110.6	-	98.9	97.9	102.6
Solvent		97.7	102.1	105.2	-	87.7	105.8	96.7

Section	Height (ft.)	Pulse Frequency (cycles/min.)
IIA Extraction	8.5	58
IIA Scrub	8	58
IIB Partitioning	8	58
IIB Scrub	8	58
IIC	12	73

All plates on 2" spacing, 1/8" diameter holes, 23% free area/plate IIA and IIB streams pulsed. IIC column pulsed directly.

Fluxmeter #2

Stream	Composition	Relative Volume
IIA	1.35 M URE, 2.0M HNO <sub>3</sub> , 0.02M NaNO <sub>2</sub>	100
IIAS	3.0 M HNO <sub>3</sub>	67
IIAX	30% Tributyl Phosphate in Amoco 123-15	333
IIBS	30% Tributyl Phosphate in Amoco 123-15	89
IIBX	0.02 M NaF - 0.6 M HNO <sub>3</sub>	89
IICX	Demineralized Water	665

\* CX/BU strip ratio 5/2

\*\*Flooded in IIA Column

Table 9  
PUREX FLOWSHEET #2 COLD SECOND U OXIDE PULSE COLUMN RUNS

Run No.	ITCP-1	ITCP-1a	ITCP-2 <sup>a</sup>	ITCP-2a**	ITCP-2	ITCP-3a	ITCP-4
Kg metal/day	50	50	75	100	100	125	150
Duration (hours)	52	48	21	...	37	20	28
Gals/hr/ft 2 (both phases)							
IIA Extraction	365	365	550	730	730	910	1095
IIA Scrub	300	300	450	600	270	340	410
IIB Partitioning	260	260	390	520	520	650	780
IIB Scrub	125	125	190	250	250	310	375
IIC	205	205	430	410	410	510	615
Column Diameter (in IPS)							
IIA Extraction	2	2	2	2	2	2	2
IIA Scrub	2	2	2	2	3	3	3
IIB Partitioning	2.5	2.5	2.5	2.5	2.5	2.5	2.5
IIB Scrub	2	2	2	2	2	2	2
IIC	4	4	4	4	4	4	4
Pulse Amplitudes in Column (in.)							
IIA Extraction	0.92	0.92	0.92	0.77	0.77	0.77	0.92
IIA Scrub	0.92	0.92	0.92	0.77	0.34	0.34	0.41
IIB Partitioning	0.64	0.64	0.86	0.86	0.86	0.43	0.54
IIB Scrub	0.92	0.92	1.23	1.23	1.23	0.62	0.77
IIC	0.42	0.56	0.56	0.56	0.56	0.56	0.56
% U Losses (Flowing Stream)							
Column IIA	0.003	0.002	0.011	-	0.011	0.008	0.008
Column IIB	0.001	0.004	<0.0001	-	0.007	0.005	0.002
Column IIC	6.62	1.95	0.12	-	0.33	0.10	0.52
% U Losses (Composite)							
Column IIA	0.42	0.001	0.50	-	0.22	0.007	1.35
Column IIB	0.004	0.007	0.015	-	0.12	0.016	0.017
Column IIC	6.15	1.46	0.09	-	2.39	0.61	7.02

% Material Balance (Comp.)					
U	-	100.6	-	99.7	102.1
HNO <sub>3</sub>	-	91.0	-	89.8	97.7
Solvent	-	115.2	-	102.2	107.6
					107.5
					94.7
					101.9

Section Diameter (In. IPS)

IIA Extraction 2

IIA Scrub 2

IIIB Partitioning 3

IIIB Scrub 3

IIC 4

# FLOWSHEET #2

Stream	Composition	Relative Volume
IIAF	1.35 M UNH, 2.0 N HNO <sub>3</sub> , 0.02 M NaNO <sub>2</sub>	100
IIAS	3.0 N HNO <sub>3</sub>	67
IIAX	30% Tributyl Phosphate in Amoco 123-15	333
IIBS	30% Tributyl Phosphate in Amoco 123-15	89
II BX	0.02 M NaF - 0.6 N HNO <sub>3</sub>	89
IICX	Demineralized Water	665

All columns packed with 1/4" x 3/8" Raschig Rings

\* Flooded in IIA Column

Table 7

PUREX FLOWSHEET #2 COLD SECOND CYCLE PACKED COLUMN RUNS

Run No.	IIC-1	IIC-2	IIC-3*	IIC-4	IIC-5	IIC-6
Kg metal/day	25	37.5	50	15	15	15
Duration, hours	72	80	27	49	50	24
Gals/hr/ft <sup>2</sup> (both phases)						
IIA Extraction	175	260	350	105	105	105
IIA Scrub	155	230	310	95	95	95
IIB Partitioning	75	110	150	45	45	45
IIB Scrub	30	45	60	20	20	20
IIC	105	160	210	60	70	95
Contacting Height (ft.)						
IIA Extraction	24	24	24	17	17	17
IIA Scrub	14	14	14	21	21	21
IIB Partitioning	28	28	28	28	28	28
IIB Scrub	13	13	13	13	13	13
IIC	22	22	22	37	37	37
CX/BU Ratio	3/2	3/2	3/2	3/2	2/1	3/1
% U Losses (flowing stream)						
Column IIA	0.004	0.014	-	0.010	0.007	0.028
Column IIB	0.002	0.007	-	0.018	0.007	0.012
Column IIC	6.8	10.5	-	0.25	0.040	0.055
% U Losses (Composite)						
Column IIA	-	2.95	-	0.019	0.003	0.007
Column IIB	-	0.002	-	0.012	0.010	0.007
Column IIC	-	7.6	-	0.20	0.061	0.068
% Material Balance (flowing stream)						
U	-	94.6	-	89.5	88.5	102.9
		07.0		06.5	03.8	09.8

Table 6

PUREX FLOWSHEET #2 COLD FIRST CYCLE PACKED COLUMN RUNS

Run No.	IC-1	IC-2	IC-3	IC-4	IC-5a	IC-5b
Kg. Metal/day	37.5	37.5	50	50	25	50
Duration (Hours)	72	81	41	46	12	12
Gals/hr/ft <sup>2</sup> (both phases)						
IA Extraction	120	120	160	160	80	160
IA Scrub	105	105	140	140	70	140
IC	160	160	195	210	105	210
Contacting Height (ft.)						
IA Extraction	20	20	20	20	20	20
IA Scrub	20	20	20	20	20	20
IC	22	22	22	22	39	39
Deviation from Flowsheet #2	None	0.02N HNO <sub>3</sub> CX	None	No CS Diluent	None	None
% U Losses (Flowing Stream)						
Column IA	0.002	0.025	0.035	0.57	0.001	0.004
Column IC	21.4	27.3	20.6	14.8	2.03	14.3
% U Losses (Composite)						
Column IA	0.34	0.18	0.28	0.78	1.49	0.21
Column IC	23.2	26.4	19.9	13.0	3.60	9.87
% Material Balance (Flowing stream)						
U	99.6	106.0	98.7	105.7	104.5	104.7
HNO <sub>3</sub>	95.4	101.4	102.0	109.4	103.8	83.5
% Material Balance (Composite)						
U	110.4	99.1	93.5	99.6	97.7	86.5
HNO <sub>3</sub>	98.3	97.9	103.3	106.7	59.2	81.5
Solvent	122.3	108.6	107.5	101.4	117.7	100.4

Section	Diameter (in. IPS)
IA Extraction	3
IA Scrub	3
IC	4

All columns packed with 1/4" x 3/8" Raschig Rings

Flowsheet #2

Stream	Composition	Relative Volume
IAF	1.35 M UNH, 2.0 N HNO <sub>3</sub> , 0.02 M NaNO <sub>2</sub>	100
IAS	3.0 N HNO <sub>3</sub>	67
IAX	30% Tributyl Phosphate in Amsco 123-15	333
ICS	30% Tributyl Phosphate in Amsco 123-15	89
ICX	Demineralized Water	665



PUREX FLOWSHEET #2 COLD FIRST CYCLE PULSE COLUMN RUNS

Run No.	ICP-1*	ICP-2*
Kg. Metal/Day	50	50
Duration (Hours)	41	24
Pulse Amplitude in Column (in.) IA IC	0.92 0.64	0.92 0.86
% U Losses (Flowing Stream) Column IA Column IC	0.002 19.4	0.003 2.50
% U Losses (Composite) Column IA Column IC	0.002 17.0	0.004 2.94
% Material Balance (Flowing Stream) U HNO <sub>3</sub>	103.0 102.0	97.8 101.4
% Material Balance (Composite) U HNO <sub>3</sub> Solvent	103.4 94.0 102.4	102.7 103.1 115.7

Section	Height (ft)	Diameter (in. IPS)	Pulse Frequency (Cycles/min.)	Gals/hr/ft <sup>2</sup> (both phases)
IA Extraction	8.5	2	58	365
IA Scrub	8	2	58	300
IC	8	2.5	58	460

All plates on 2" spacing, 1/8" diameter holes, 23% free area/plate  
IAX and ICU streams pulsed

\* No ICS diluent stream employed

Flowsheet #2

<u>Stream</u>	<u>Composition</u>	<u>Relative Volume</u>
IAF	1.35 M UNH, 2.0 N HNO <sub>3</sub> , 0.02M NaNO <sub>2</sub>	100
IAS	3.0 N HNO <sub>3</sub>	67
IAX	30% Tributyl Phosphate in Amsco 123-15	333
ICX	Demineralized Water	532

Table 10

PUREX FLOWSHEET #1 HOT FIRST CYCLE PULSE COLUMN RUNS

Run	IHP-1	IHP-2	IHP-3	IHP-4	IHP-5
Feed $\alpha$ Activity (cts/min/ml)	$6.7 \times 10^4$	$1.8 \times 10^5$	$1.6 \times 10^6$	$1.2 \times 10^6$	$9.4 \times 10^5$
Feed $\beta$ Activity (cts/min/ml)	$7.2 \times 10^6$	$1.4 \times 10^7$	$1.8 \times 10^8$	$2.6 \times 10^8$	$2.6 \times 10^8$
Feed $\gamma$ Activity (mv/ml)	$1.9 \times 10^3$	$4.5 \times 10^3$	$1.1 \times 10^5$	$2.2 \times 10^5$	$2.2 \times 10^5$
Throughput (Kg metal/day)	125	125	125	125	75
Duration (hours)	32	32	35	36	61
Gals/hr/ft. <sup>2</sup> (both phases)					
IA Extraction	910	910	910	910	555
IA Scrub	340	340	340	340	460
IB Partitioning	650	650	650	650	390
IB Scrub	310	310	310	310	190
IBs	195	195	195	195	120
IC	510	510	510	510	310
Diameter (in. IPS)					
IA Extraction	2	2	2	2	2
IA Scrub	3	3	3	3	2
IB Partitioning	2.5	2.5	2.5	2.5	2.5
IB Scrub	2	2	2	2	2
IBs	2	2	2	2	2
IC	4	4	4	4	4
Contacting Height (ft.)					
IA Extraction	7.5	7.5	7.5	7.5	7.5
IA Scrub	12.5	12.5	12.5	12.5	12.5
IB Partitioning	11.5	11.5	11.5	11.5	11.5
IB Scrub	6	6	6	6	6
IBs	15	15	15	15	15
IC	12	12	12	12	12
Pulse Frequency (c.p.m.)					
IA Column	58	58	58	58	58
IB Column	58	58	58	58	58
IC Column	73	73	73	73	73
Pulse Amplitude (in.)					
IA Extraction	0.84	0.92	0.92	0.92	0.92
IA Scrub	0.37	0.41	0.41	0.41	0.92
IB Partitioning	0.53	0.53	0.53	0.53	0.75
IB Scrub	0.77	0.77	0.77	0.77	1.08
IC	0.56	0.49	0.42	0.49	0.56
% U Losses (flowing stream)					
Column IA	0.07	0.06	0.07	0.07	0.06
Column IB	0.06	0.10	0.09	0.03	0.0007
Column IC	0.03	0.024	0.51	0.42	0.50
% U Losses (composite)					
Column IA	1.10	0.06	0.07	1.64	0.12
Column IB	0.21	0.15	0.13	0.21	0.10
Column IC	0.04	0.04	0.27	0.40	0.56

(Table continued next page)

Table 10

PUREX FLOWSHEET #1 HOT FIRST CYCLE PULSE COLUMN RUNS (continued)

Run	IHP-1	IHP-2	IHP-3	IHP-4	IHP-5
% Pu Losses (flowing stream)					
Column IA	1.21	0.29	1.19	0.90	0.62
Column IB	3.90	2.42	1.37	1.29	0.38
% Pu Losses (composite)					
Column IA	1.68	0.23	1.14	4.98	1.51
Column IB	3.12	1.64	1.50	1.71	0.48
% Material Balance (flowing stream)					
U	102.3	106.7	105.3	109.0	91.3
HNO <sub>3</sub>	105.2	116.7	99.7	92.3	87.0
% Material Balance (composite)					
Pu	106.2	86.8	93.2	102.9	103.9
U	89.0	95.4	104.4	98.3	93.9
HNO <sub>3</sub>	109.1	117.6	103.1	102.8	94.4
Solvent	82.2	93.9	94.8	99.7	98.9
IAP Decontamination Factor					
*Gross $\beta$	$2.8 \times 10^3$	$3.4 \times 10^3$	580	820	$2.9 \times 10^3$
Ru $\beta$	220	120	120	190	150
Zr $\beta$	43	66	62	120	$3.1 \times 10^3$
Cb $\beta$	790	-	320	870	$1.6 \times 10^3$
TRE $\beta$	$3.5 \times 10^3$	$5.7 \times 10^3$	$6.7 \times 10^3$	$1.1 \times 10^4$	$2.0 \times 10^5$
Gross $\gamma$	450	290	130	220	570
IBsP Decontamination Factor					
*Gross $\beta$	$8.1 \times 10^3$	$9.5 \times 10^3$	830	$1.2 \times 10^3$	$1.1 \times 10^4$
Ru $\beta$	$1.8 \times 10^3$	$1.1 \times 10^3$	$1.2 \times 10^3$	$1.2 \times 10^3$	$1.4 \times 10^3$
Zr $\beta$	50	69	55	110	$3.5 \times 10^3$
Cb $\beta$	-	320	310	420	$3.0 \times 10^3$
TRE $\beta$	$7.0 \times 10^3$	$3.8 \times 10^4$	$4.9 \times 10^3$	$8.2 \times 10^3$	$7.3 \times 10^4$
Gross $\gamma$	850	680	160	270	$3.8 \times 10^3$
ICU Decontamination Factor					
*Gross $\beta$	$1.9 \times 10^4$	$1.6 \times 10^4$	$6.5 \times 10^3$	$1.5 \times 10^4$	$2.3 \times 10^4$
Ru $\beta$	610	840	660	$1.1 \times 10^3$	$1.1 \times 10^3$
Zr $\beta$	160	$2.4 \times 10^3$	$4.2 \times 10^4$	$7.6 \times 10^3$	$1.4 \times 10^5$
Cb $\beta$	-	660	400	790	$1.8 \times 10^3$
TRE $\beta$	$2.8 \times 10^5$	$1.6 \times 10^5$	$7.4 \times 10^5$	$1.3 \times 10^6$	$3.1 \times 10^6$
Gross $\gamma$	500	$1.8 \times 10^3$	990	$1.7 \times 10^3$	$4.0 \times 10^3$
Ratio IBsX/IBP	1/4	1/4	1/4	1/2	1/2

\* Gross  $\beta$  D.F. corrected for  $UX_1 + UX_2$  where correction was  $> 10\%$ .

(Table continued next page)

Table 10

PUREX FLOWSHEET #1 HOT FIRST CYCLE PULSE COLUMN RUNS (continued)

Flowsheet #1

<u>Stream</u>	<u>Composition</u>	<u>Relative Volume</u>
IAF	1.35M UNH, 2.0N HNO <sub>3</sub> , 0.02M NaNO <sub>2</sub>	100
IAS	3.0N HNO <sub>3</sub>	67
IAX	30% Tributyl Phosphate in Amsco 123-15	333
IBS	30% Tributyl Phosphate in Amsco 123-15	89
IBX	0.03M Fe(NH <sub>2</sub> SO <sub>3</sub> ) <sub>2</sub> , 0.6N HNO <sub>3</sub>	89
IBsX	Amsco 123-15	44
ICX	Demineralized H <sub>2</sub> O	665

All pulse plates on 2" spacing, 1/8" diameter holes, 23% free area/plate. IBS column packed with Raschig Rings (1/2"x1/2" for Run IHP-5, 1/4"x1/4" for all other runs).

Table 11

PUREX FLOWSHEET #1 HOT SECOND URANIUM CYCLE PULSE COLUMN RUNS

Run	IIHP-1	IIHP-3**	IIHP-4	IIHP-5
Feed $\alpha$ activity (cts/min./ml)	$3.1 \times 10^3$	$2.2 \times 10^4$	$1.4 \times 10^4$	$3.9 \times 10^3$
Feed $\beta$ activity (cts/min./ml)	$1.6 \times 10^4$	$2.6 \times 10^4$	$1.9 \times 10^4$	$1.6 \times 10^4$
Feed $\text{Fe}(\text{NH}_2\text{SO}_3)_2$ Molarity	0.03	0.03	0.02	0.02
Scrub $\text{Fe}(\text{NH}_2\text{SO}_3)_2$ Molarity	0.00	0.00	0.02	0.02
Throughput (Kg metal/day)	125	125	100	100
Duration (hours)	28	21	27	51
Gals/hr/ft. <sup>2</sup> (both phases)				
ID Extraction	910	910	730	730
ID Scrub	340	340	270	270
IE	410	410	520	520
Diameter (in. IPS)				
ID Extraction	2	2	2	2
ID Scrub	3	3	3	3
IE	4	4	4	4
Contacting Height (ft.)				
ID Extraction	8.5	8.5	8.5	8.5
ID Scrub	8	8	8	8
IE	15	15	15	15
Pulse Frequency (c.p.m.)				
ID Column	58	58	58	58
IE Column	73	73	73	73
Pulse Amplitude (in.)				
ID Extraction	1.08	1.08	1.00	1.00
ID Scrub	0.48	0.48	0.44	0.44
IE	0.49	0.49	0.56	0.56
% U Losses (flowing stream)				
Column ID	0.003	5.94	0.01	0.003
Column IE	2.00	0.35	0.88	1.09
% U Losses (composite)				
Column ID	2.12	2.80	0.94	0.002
Column IE	-	-	-	0.74
% Material Balance (flowing stream)				
U	79.3	94.3	66.7	119.4
HNO <sub>3</sub>	96.3	89.5	78.0	92.9
% Material Balance (composite)				
U	81.4	107.8	91.8	74.6
HNO <sub>3</sub>	79.0	89.6	90.1	82.3
Solvent	117.7	96.9	102.5	104.1
IDU Decontamination Factor				
*Gross $\beta$	-	25	3	2
Ru $\beta$	3	17	7	2
Zr $\beta$	1	2	1	1
Cb $\beta$	-	-	-	-
TRE $\beta$	2	5	-	-

\* Corrected for  $\text{UX}_1 + \text{UX}_2$

\*\* 27% TBP solvent

(Table continued next page)

Table 11

PUREX FLOWSHEET #1 HOT SECOND URANIUM CYCLE PULSE COLUMN RUNS (continued)

Run	IIHP-1	IIHP-3	IIHP-4	IIHP-5
IEU Decontamination Factor				
*Gross $\beta$	-	56	15	9
Ru $\beta$	-	95	39	20
Zr $\beta$	-	4	1	2
Cb $\beta$	-	57	42	49
TRE $\beta$	-	5	1	1
Pu $\alpha$	5	6	48	26
**Uranium D.F. through two cycles				
Gross $\beta$	$>10^4$	$2.1 \times 10^5$	$3.2 \times 10^5$	$3.4 \times 10^5$
Ru $\beta$	-	$6.9 \times 10^4$	$4.1 \times 10^4$	$2.0 \times 10^4$
Zr $\beta$	-	$1.6 \times 10^5$	$1.5 \times 10^5$	$2.3 \times 10^5$
Cb $\beta$	-	$3.6 \times 10^4$	$2.6 \times 10^4$	$1.1 \times 10^5$
TRE $\beta$	-	$1.0 \times 10^7$	$1.3 \times 10^6$	$2.6 \times 10^6$
Gross $\gamma$	-	$3.7 \times 10^3$	$4.2 \times 10^4$	$8.3 \times 10^4$
Pu in Uranium Product (p.p.b.)	20	30	13	7
F.P. $\beta$ in Uranium Product (% of old natural U)	30	2	2	1

All pulse plates on 2" spacing, 1/8" diameter holes, 23% free area/plate.

Flowsheet #1

<u>Stream</u>	<u>Composition</u>	<u>Volume</u>
IDF	1.35M UNH, 2.0N HNO <sub>3</sub> , 0.02M Fe(NH <sub>2</sub> SO <sub>3</sub> ) <sub>2</sub>	100
IDS	0.02M Fe(NH <sub>2</sub> SO <sub>3</sub> ) <sub>2</sub> , 3.0N HNO <sub>3</sub>	67
IDX	30% Tributyl Phosphate in Amsco 123-15	333
IEX	Demineralized H <sub>2</sub> O	532

\* Corrected for UX<sub>1</sub> + UX<sub>2</sub>

\*\* Corrected for cold UNH from startup and shutdown

Table 12

## PUREX FLOWSHEET #1 HOT SECOND PLUTONIUM CYCLE PACKED COLUMN RUNS

Run	III HP-1	III HP-2a	III HP-2b	III HP-3a	III HP-3b	III HP-4a	III HP-4b	III HP-5a	III HP-5b
Feed $\alpha$ activity (cts/min/ml)	2.4x10 <sup>5</sup>	6.2x10 <sup>5</sup>	6.2x10 <sup>5</sup>	7.1x10 <sup>5</sup>	7.1x10 <sup>5</sup>	4.6x10 <sup>5</sup>	4.6x10 <sup>5</sup>	5.6x10 <sup>5</sup>	4.4x10 <sup>5</sup>
Feed $\beta$ activity (cts/min/ml)	4.7x10 <sup>4</sup>	6.7x10 <sup>4</sup>	5.7x10 <sup>4</sup>	1.0x10 <sup>6</sup>	1.0x10 <sup>6</sup>	1.2x10 <sup>6</sup>	1.2x10 <sup>6</sup>	3.6x10 <sup>4</sup>	1.3x10 <sup>5</sup>
Feed $\gamma$ activity (mv/ml)	43	43	43	1.8x10 <sup>3</sup>	1.8x10 <sup>3</sup>	1.9x10 <sup>3</sup>	1.9x10 <sup>3</sup>	46	143
Duration (hours)	21	18	22	19	18	18	8	28	37
Gals/hr/ft. <sup>2</sup> (both phases)									
Column IIA	300	300	250	300	300	200	200	200	200
Column IIB	225	225	200	225	225	125	125	125	125
Diameter (in. IPS)									
Column IIA	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Column IIB	2	2	2	2	2	2	2	2	2
Column IIBs	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5	1.5
Contacting Height (ft.)									
IIA Extraction	30	30	30	26	16	30	30	16	16
IIA Scrub	10	10	10	14	24	10	10	24	24
IIB	40	40	40	40	40	40	40	40	40
IIBs	27	27	27	27	27	27	27	27	27
Flow Ratio IIAS:IIAF:IIAX	2:3:10	2:3:10	2:3:7.5	2:3:10	2:3:10	2:6:10	2:6:10	2:6:12	2:6:12
% Pu Losses									
Column IIA	0.47	0.13	0.04	0.007	0.007	0.02	0.02	0.09	0.03
Column IIB (H <sub>2</sub> O IIBX)	0.51	0.46	0.75	3.42	3.82	0.86	-	0.42	0.55
Column IIB (0.1N HNO <sub>3</sub> IIBX)	-	-	-	-	-	-	0.27	0.31	0.14
Column IIBs	0.04	0.02	0.004	0.001	0.002	0.003	0.006	0.015	0.003
Pu Material Balance (%)**	101.8	-	110.4	-	85.2	-	84.2	118.7	112.3
IIAP Decontamination Factor									
*Gross $\beta$	1	12	10	269	296	409	441	33	135
Ru $\beta$	2	2	2	52	53	90	87	8	96
Zr $\beta$	13	3	17	351	581	451	502	81	163
TRE $\beta$	-	-	-	-	141	-	-	-	-

\* Corrected for UX<sub>1</sub> + UX<sub>2</sub>

\*\* For a and b runs together

(Table continued next page)

Table 12

PUREX FLOWSHEET #1 HOT SECOND PLUTONIUM CYCLE PACKED COLUMN RUNS (continued)

Run	III HP-1	III HP-2a	III HP-2b	III HP-3a	III HP-3b	III HP-4a	III HP-4b	III HP-5a	III HP-5t
<b>IIBsP Decontamination Factor</b>									
*Gross $\beta$	54	44	72	319	478	441	524	94	120
Ru $\beta$	42	30	41	58	93	210	330	90	325
Zr $\beta$	42	57	91	446	626	433	494	59	102
TRE $\beta$	-	36	74	332	621	2.1x10 <sup>3</sup>	3.7x10 <sup>3</sup>	891	41
Gross $\gamma$	-	22	17	338	423	203	225	43	160
<b>Plutonium D.F. through two cycles</b>									
*Gross $\beta$	4.4x10 <sup>5</sup>	4.2x10 <sup>5</sup>	6.9x10 <sup>5</sup>	2.6x10 <sup>5</sup>	3.9x10 <sup>5</sup>	5.3x10 <sup>5</sup>	6.3x10 <sup>5</sup>	1.0x10 <sup>6</sup>	1.3x10 <sup>6</sup>
Ru $\beta$	7.6x10 <sup>4</sup>	3.3x10 <sup>4</sup>	4.5x10 <sup>4</sup>	7.0x10 <sup>4</sup>	1.1x10 <sup>5</sup>	2.5x10 <sup>5</sup>	3.9x10 <sup>5</sup>	1.2x10 <sup>5</sup>	4.3x10 <sup>5</sup>
Zr $\beta$	2.1x10 <sup>3</sup>	3.9x10 <sup>3</sup>	6.2x10 <sup>3</sup>	2.5x10 <sup>4</sup>	3.5x10 <sup>4</sup>	4.7x10 <sup>4</sup>	5.4x10 <sup>4</sup>	2.1x10 <sup>5</sup>	3.6x10 <sup>5</sup>
TRE $\beta$	-	1.4x10 <sup>6</sup>	2.9x10 <sup>6</sup>	1.6x10 <sup>6</sup>	3.0x10 <sup>6</sup>	1.7x10 <sup>7</sup>	3.0x10 <sup>7</sup>	6.5x10 <sup>7</sup>	3.0x10 <sup>6</sup>
Gross $\gamma$	-	1.5x10 <sup>4</sup>	1.2x10 <sup>4</sup>	5.4x10 <sup>4</sup>	6.7x10 <sup>4</sup>	5.5x10 <sup>4</sup>	6.1x10 <sup>4</sup>	1.6x10 <sup>5</sup>	5.9x10 <sup>5</sup>

\*Corrected for UX<sub>1</sub> UX<sub>2</sub>

Columns packed with 1/4 inch by 3/8 inch Raschig rings.

Stream	Flowsheet #1	
	Composition	Relative Volume
IIF	5.0N HNO <sub>3</sub>	100
IIS	5.0N HNO <sub>3</sub>	67
IIX	10% Tributyl Phosphate in Amsco 123-15	333
IIBX	Demineralized H <sub>2</sub> O	333
IIBsX	Amsco 123-15	83



Appendix

Calculation of HETS

The number of theoretical stages was determined graphically by counting the number of steps between the operating and equilibrium lines on a plot of organic vs. aqueous phase uranium molarities. The position of the equilibrium curves is influenced strongly by the nitric acid concentration as is shown in KAPL-461, Figure 4. Nitric acid reflux within the columns was assumed to be identical for the columns as for the mixer-settlers from which the data were drawn. Figures 20, 21, and 22 of KAPL-461 were employed since none of the runs calculated yielded more than five stages for the IA extraction or IB scrub sections. Operating lines were determined from Figures 12, 13 of the present report.

Calculation of HTU

Colburn defined NTU by:

$$NTU = \int_{y_1}^{y_2} \frac{(1-y) y dy}{(1-y) (y-y^*)} \quad (1)$$

The term  $(1-y)_f / (1-y)$  was neglected as it is 1.00 for the IA extraction and IB scrub sections, and varies from 1.00 to 1.01 in the IC column. Therefore we can simplify to:

$$NTU = \int_{y_1}^{y_2} \frac{dy}{y-y^*} \quad (2)$$

where  $y$  is the concentration of uranium in one phase and  $y^*$  is the equilibrium concentration in the same phase corresponding to the other phase. Although equation (2) is based on mol fraction units the use of g/l units has a negligible

Calculation of HTU (continued)

effect since the concentrations are less than 1M. Overall aqueous phase HTU's for the IA extraction and IB scrub sections and overall organic phase HTU for the IC column were calculated. Equation (2) was integrated using values of  $y$  and  $y^*$  from the same equilibrium and operating lines used for the HETS calculation. In the equation for the IA extraction section

$$(NTU)_{OA} = 3.22 + 1.03 \ln \frac{9.24 + 0.0294 y_1}{y_1} \quad (3)$$

$y_1$  is the IAW uranium concentration in grams per liter and the constant 3.22 represents the number of transfer units required to reduce the aqueous uranium molarity from 0.86, the calculated value at the feed point to 0.04. Over this range the equilibrium line is curved and a graphic integration was performed. For  $y$  below 0.04M the equilibrium line is straight and the integration was performed analytically. The residual uranium in the IAW was taken into account and has only a small effect. The integrated form for the IB scrub section is:

$$(NTU)_{OA} = 1.05 \ln \frac{28.3 + 0.05 y_1}{y_1} \quad (4)$$

where  $y_1$  is the IBP uranium concentration in grams per liter. The equilibrium line is straight and the analytical method was used. A correction was made for residual uranium in the IBP. The aqueous uranium concentration at the feed point was calculated as 0.125M. For the IC column the curved equilibrium line makes a graphical solution simplest. A plot of the integrated results is given in Figure

10 as  $(NTU)_{OO}$  vs. uranium content in the ICW.

Calculation of HTU (continued)

The equilibrium curve has much less effect on HTU than on HETS since if  $y$  is much larger than  $y^*$  then  $y^*$  can be neglected and equation (2) becomes

$$NTU = \int_{y_1}^{y_2} \frac{dy}{y} \quad (5)$$

If the equilibrium line for twelve IA extraction stages (KAPL-461, Figure 17) were used instead of the curve for five stages the values of NTU would decrease less than 1 percent. If the curve for ten IB scrub stages (KAPL-461, Figure 18) were used instead of that for five stages the NTU values would increase about 7 percent. The use of the ten stage instead of the five stage equilibrium line about would increase the number of IB scrub stages by 30 percent.

When the equilibrium and operating lines are both straight, for overall aqueous HTU:

$$\frac{HTU}{HETS} = \frac{(m/R) + 1}{\ln(m/R)} \quad (6)$$

where  $m$  is the slope of the equilibrium curve and  $R$  is the slope of the operating line. The equation applied to the IB scrub section with  $m = 20$  and  $R = 1$  yielded  $HETS = 3.15$  HTU which is a close check upon the values computed from the runs.

The data from the IA and ID columns for runs with Flowsheet No. 1 and the IA and IIA columns for runs with Flowsheet No. 2 were used in the IA extraction calculations. The Flowsheet No. 1 - IB column data and the Flowsheet No. 2 - IIB column data were used for IB scrub calculations, while the IC strip calculations were made using Flowsheet No. 1 - IC column losses and Flowsheet No. 2 - IIC column losses. The presence of ferrous sulfamate in column ID in

Calculation of HTU (continued)

Run IIS-1 and of sodium fluoride in the IIB column in the IIC and IICP runs was considered to have a negligible effect on the uranium transfer. Data for the IC column of the IC and ICP runs and of the IE column from Run IIS-1 were not calculated since equilibrium data were not available for the nitric acid concentration range involved.

Table 13

PUREX URANIUM HTU AND HETS RESULTS

<u>Packed Columns</u>						
<u>Run No.</u>	<u>IA Extraction</u>		<u>IB Scrub</u>		<u>IC Strip</u>	
	HTU <sub>OA</sub> (ft.)	HETS(ft.)	HTU <sub>OA</sub> (ft.)	HETS(ft.)	HTU <sub>OO</sub> (ft.)	HETS(ft.)
IC-1	1.77	5.9	-	-	-	-
2	2.33	7.4	-	-	-	-
3	2.41	7.7	-	-	-	-
4	-	-	-	-	-	-
5a	1.95	6.5	-	-	-	-
5b	1.99	6.5	-	-	-	-
IIC-4	1.80	5.9	2.03	6.5	4.65	7.9
5	1.73	5.7	1.81	5.9	-	8.4*
6	2.01	6.5	1.93	6.2	-	12.3*
IS-1	1.97	6.5	1.43	4.6	8.30	11.1
2	1.62	5.4	1.21	3.8	-	16.2*
3	1.48	5.0	1.27	4.1	7.47	10.3
4	1.61	5.4	-	-	-	13.4*
IIS-1,1a	1.82	5.9	-	-	-	-
Average	1.88	6.2	1.61	5.2	6.81	9.8
<u>Pulse Columns</u>						
ICP-1	0.75	2.5	-	-	-	-
-2	0.80	2.7	-	-	-	-
IICP-1	0.80	2.7	0.91	2.9	2.75	3.5
1a	0.79	2.7	1.02	3.2	2.09	3.0
2	0.90	2.9	0.66	2.1	-	3.9*
2a	-	-	-	-	-	-
3	0.90	2.9	1.11	3.5	1.58	2.6
3a	0.87	2.8	1.03	3.3	1.30	2.4
4	0.86	2.8	0.92	3.0	1.72	2.7
ISP-1	0.76	2.5	-	-	2.45	3.3
2	0.95	3.1	1.21	3.8	2.16	3.1
3	0.87	2.8	-	-	1.92	2.9
4	0.85	2.8	0.80	2.6	0.95	1.7
5	-	-	0.87	2.8	2.28	3.2
6	1.07	3.4	1.46	4.7	-	-
7	0.93	3.0	1.05	3.3	-	-
8	0.80	2.6	1.02	3.3	0.96	1.7
9	0.88	2.9	0.93	3.0	1.09	1.9
10	0.92	3.0	-	-	2.28	3.2
11	0.99	3.1	0.90	2.9	1.12	2.0
12	0.88	2.9	0.83	2.6	1.15	2.1
Average	0.87	2.8	0.98	3.1	1.72	2.6

\* Not included in average because ICX/IBU strip ratio differed from 1.5.

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Table 14

PUREX COLD IAW EVAPORATION AND ACID RECOVERY

	Run ISP-8	Run ISP-9
Average Recovered Acid Strength (% $\text{HNO}_3$ )	63.1	62.0
Volume of Acid Recovered (liters)	43	56
Average Condensate Acidity (% $\text{HNO}_3$ )	0.3	0.2
% Acid Lost to Condensate	1.7	1.3
Material Balance (% of entering IAW $\text{HNO}_3$ in acid and condensate catch tanks)	61.7	60.3
IAW Volume Reduction Factor	5.8	4.9
De-entrainment Factor for U (evaporator to recovered acid)	$2 \times 10^4$	$2 \times 10^2$
De-entrainment Factor for U (evaporator to condensate)	$2 \times 10^3$	$6 \times 10^2$
Final Evaporator acidity (N)	8.63	9.34
Ratio Reflux to Condensed Distillate	0.30	0.27
Run Duration, hours	24	24
IAW Acid Concentration (N)	1.84	2.14

Fractionating Column

<u>Section</u>	<u>Diameter (in IPS)</u>	<u>Height (ft.)</u>	<u>Packing</u>
Stripping	4	9.5	7/8" x 7/8" Raschig Rings
Rectifying	6	5	7/8" x 7/8" Raschig Rings

Feed - Vapor from evaporator  
Column Vapor Velocity - 0.1 ft/sec

Table 15

PUREX RUN IHP-5 HOT IAW EVAPORATION AND ACID RECOVERY

IAW Average Acidity (N)	2.14
IAW Feed Rate (gals/hr.)	3.75
Average Recovered Acid Strength (% $\text{HNO}_3$ )	56.4
Volume of Acid Recovered (Liters)	158
Average Condensate Acidity (%)	0.14
% Acid Lost to Condensate	1.9
Material Balance (% of entering IAW $\text{HNO}_3$ in acid and condensate catch tanks)	109
IAW Volume Reduction Factor (during run)	7.1
IAW Volume Reduction Factor (after final boildown)	20.8
IAW Gross $\beta$ (cts/min/ml)	$1.7 \times 10^8$
Evaporator Gross $\beta$ (after volume reduction during run)	$1.1 \times 10^9$
Gross $\beta$ D.F. for Recovered Acid	$4.0 \times 10^5$
Gross $\beta$ D.F. for Condensate	$>10^6$
Evaporator $\text{HNO}_3$ Normality (after volume reduction during run)	8.82
Run Duration (continuous boildown), hours	61
Ratio Reflux to Condensed Distillate	0.34

Column Arrangement same as for cold runs (see previous table).

Table 16

Composition and Volume of Purex Primary Waste Streams

Basis: Full ORNL Level  
Feed Volume-100

<u>Stream</u>	<u>Composition</u>	<u>Relative Volume</u>
ORNL Coating Removal	2.7 M $\text{NaNO}_3$ 2.8 M $\text{NaOH}$ 0.86 M $\text{Al}$ U - 0.1 g/l Pu - $10^3$ c/m/ml Gross $\beta$ - $10^6$ c/m/ml	29
Hanford Coating Removal	2.7 M $\text{NaNO}_3$ 4.0 M $\text{NaOH}$ 1.32 M $\text{Al}$ U - 0.1 g/l	
Coating Removal Rinse No. 1	$\text{H}_2\text{O}$	42
Coating Removal Rinse No. 2	$\text{H}_2\text{O}$	25
Filter Cake Wash No. 1	$\text{H}_2\text{O}$ U - 10 g/l Pu - $10^4$ c/m/ml Gross $\beta$ - $10^5$ c/m/ml	28
Filter Cake Wash No. 2	7.5 M $\text{NaOH}$ U - 0.05 g/l Pu - 100 c/m/ml Gross $\beta$ - $10^5$ c/m/ml	28
Filter Cake Wash No. 3	4.5 M $\text{HNO}_3$ U - 0.03 g/l Pu - 50 c/m/ml Gross $\beta$ - $1 \times 10^4$ c/m/ml	28
<u>Dissolver Off-Gas</u>	<u>Flowsheet Uncertain</u>	
IAW	2.25 M $\text{HNO}_3$ 0.012 M $\text{Na}^+$ U - 0.1 g/l Pu - $10^3$ c/m/ml Gross $\beta$ - $1.5 \times 10^8$ c/m/ml	152
ICW	30% TBP - 70% Amsco (vol.) U - 0.1 g/l Pu - 200 c/m/ml Gross $\beta$ - $10^4$ c/m/ml	423

(Table continued on next page)



Table 16

Composition and Volume of Purex Primary Waste Streams (continued)

<u>Stream</u>	<u>Composition</u>	<u>Relative Volume</u>
ICU Evap. Condensate	H <sub>2</sub> O Gross $\beta$ - 50 c/m/ml	595
IBsP Evap. Condensate	H <sub>2</sub> O Gross $\beta$ - 80 c/m/ml Gross $\alpha$ - 100 c/m/ml	71
IBsW	Amsco (123-15) Trace of TBP U - < 0.0001 g/l Pu - < 10 c/m/ml Gross $\beta$ < 50 c/m/ml	22.2
IDW	2.25 M HNO <sub>3</sub> Fe - 0.02 M U - 0.1 g/l Pu - 10 <sup>4</sup> c/m/ml Gross $\beta$ - 5x10 <sup>3</sup> c/m/ml	152
IEW	30% TBP - 70% Amsco (vol.) Gross $\beta$ - 300 c/m/ml	349
IEU Evap. Condensate	H <sub>2</sub> O U - 0.01 g/l Pu - < 20 c/m/ml Gross $\beta$ - < 10 c/m/ml	480
IIAW	3.7 M HNO <sub>3</sub> 0.09 M Fe <sup>3++</sup> Pu - < 500 c/m/ml Gross $\beta$ - 10 <sup>5</sup> c/m/ml	29.6
IIBW	10% TBP - 90% Amsco 123-15 Pu - 3x10 <sup>3</sup> c/m/ml Gross $\beta$ - 150 c/m/ml	59.2
IIBsP Evap. Condensate	0.02 M HNO <sub>3</sub> Pu - 150 c/m/ml Gross $\beta$ 50 c/m/ml	58
IIBsW	Amsco 123-15 Trace TBP	14.8
Final U Purification	Flowsheet Uncertain	
Final Pu Purification	Flowsheet Uncertain	

Pu counts at 50% geometry  
Gross Beta counts at 10% geometry

Table 17

Composition and Relative Volumes of Purex Secondary Wastes

<u>System</u>	<u>Primary Waste Source</u>	<u>Secondary Waste Composition</u>	<u>Relative Volume</u>
Solvent Recovery	ICW, IEW, IIBW	0.1 M $\text{Na}_2\text{CO}_3$	160
		Gross $\beta$ - $10^4$ c/m/ml	
		Pu - $10^3$ - c/m/ml	
		<hr/> H <sub>2</sub> O	160
Nitric Acid Recovery	IAW	0.01 M $\text{HNO}_3$	304
		Gross $\beta$ - 30 c/m/ml	
		<hr/> 8.0 M $\text{HNO}_3$	15.2
		Gross $\beta$ - $10^9$ c/m/ml	
		Pu - $10^4$ c/m/ml	
		U - 1.0 g/l	

Pu counts at 50% geometry  
 Gross beta counts at 10% geometry  
 Volumes based on 100 volumes of IAF  
 Activities based on full ORNL level

Table 18

APPROXIMATE SPECIFIC GRAVITY OF PUREX FLOWSHEET #1 STREAMS

<u>STREAM</u>	<u>Sp.G 25/25°C</u>	<u>STREAM</u>	<u>Sp.G. 25/25°C</u>
IAF	1.50	IDU	0.97
IAS	1.09	IDW	1.08
IAX	0.84	IEX	1.00
IAP	0.97	IEU	1.08
IAW	1.07	IEW	0.84
IBS	0.84	IIAF	1.19
IBX	1.04	IIAS	1.16
IBU	0.94	IIAX	0.80
IBP	1.05	IIAP	0.81
IBsX	0.78	IIAW	1.17
IBsP	1.05	IIBX	1.00
IBsW	0.78	IIBP	1.01
ICX	1.00	IIBW	0.80
ICU	1.06	IIBsX	0.79
ICW	0.84	IIBsP	1.01
IDF	1.51	IIBsW	0.79
IDS	1.10	EAW	1.25
IDX	0.84	ER	1.34